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(54) ORGANIC ELECTROLUMINESCENT ELEMENT

(57) The novel compound of the present invention has the diphenylanthracene structure at the center and a specific structure substituted with an aryl group at end portions. The organic electroluminescence device of the present invention comprises a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes

and at least one of the layers of thin films of organic compounds comprises the above novel compound. The novel compound exhibits excellent efficiency of light emission and heat resistance, has a long life and emits bluish light having excellent purity of color and the organic electroluminescence device comprises the novel compound and exhibits the same advantageous properties.

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Description**TECHNICAL FIELD**

5 [0001] The present invention relates to a novel compound which is used as a light source such as a planar light emitting member of wall televisions and a back light of displays, exhibits excellent efficiency of light emission and heat resistance, has a long life and emits bluish light having excellent purity of color and to an organic electroluminescence device utilizing the novel compound.

BACKGROUND ART

10 [0002] Electroluminescence (referred to as EL, hereinafter) devices using organic substances are expected to be used for inexpensive full color display devices of the solid light emission type which can display a large area and development thereof has been actively conducted. In general, an EL device is constituted with a light emitting layer and a pair of electrodes faced to each other at both sides of the light emitting layer. When a voltage is applied between the electrodes, electrons are injected at the side of the cathode and holes are injected at the side of the anode. The electrons are combined with the holes in the light emitting layer and an excited state is formed. When the excited state returns to the ground state, the energy is emitted as light.

15 [0003] Organic EL devices used heretofore have higher driving voltages, lower luminances of emitted light and lower efficiencies of light emission than inorganic light emitting diodes and, moreover, the properties of organic EL devices deteriorate rather rapidly. Therefore, organic EL devices have not been used for practical applications. Although the properties of recent organic EL devices have been improved remarkably, the efficiency of light emission, the heat resistance and the life are not sufficient for practical applications.

20 [0004] For example, an organic EL device using a dimer or a trimer of phenylanthracene is disclosed in Japanese Patent Application Laid-Open No. Heisei 8(1996)-12600. However, since the above compound used in the device contains two or three anthracene structures bonded through a conjugated group, the organic EL device has a small energy gap and exhibits poor purity of color in emission of blue light. Moreover, since this compound is easily oxidized, impurities tend to be contained and a problem arises with respect to purification. To overcome the above problems, preparation of organic EL devices using a compound in which the 1-position and the 9-position of anthracene are substituted with naphthalene or a compound in which m-position of phenyl group in diphenylanthracene is substituted with an aryl group have been examined. However, the prepared organic EL devices exhibit low efficiencies of light emission and cannot be used for practical applications.

25 [0005] An organic EL device using a monoanthracene derivative substituted with naphthalene is disclosed in Japanese Patent Application Laid-Open No. Heisei 11(1999)-3782. However, this device exhibits an efficiency of light emission as low as 1 cd/a and cannot be used for practical applications. An organic EL device using a compound having a phenylanthracene structure is disclosed in United States Patent No. 5,972,247. However, this device exhibits an efficiency of light emission as low as 2 cd/A and cannot be used for practical applications although the device exhibits excellent heat resistance due to the substitution with an aryl group at the m-position.

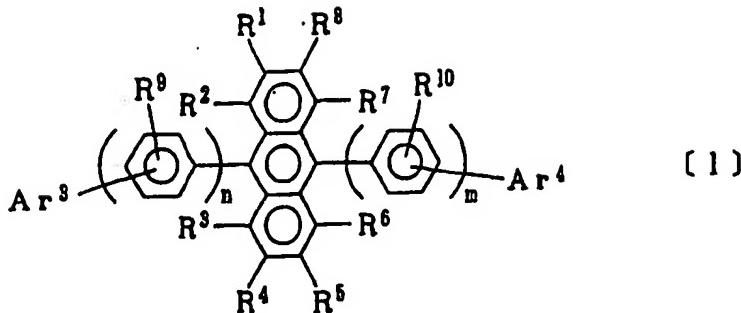
DISCLOSURE OF THE INVENTION

30 [0006] The present invention has been made to overcome the above problems and has an object of providing a novel compound which exhibits excellent efficiency of light emission and heat resistance, has a long life and emits bluish light having excellent purity of color and an organic electroluminescence device utilizing the novel compound.

35 [0007] As the result of extensive studies by the present inventors to develop a novel compound having above advantageous properties and an organic EL device using the novel compound, it was found that the object can be achieved by using a compound having the diphenylanthracene structure at the center and a specific structure substituted with an aryl group at end portions. The present invention has been completed based on the knowledge.

40 [0008] The novel compound of the present invention comprises compounds represented by the following general formulae [1], [1'] and [2] to [5].

General formula [1]:



20 wherein R¹ to R¹⁰ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

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[0009] Ar³ and Ar⁴ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 2 to 10 carbon atoms, the substituent in the above groups being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms;

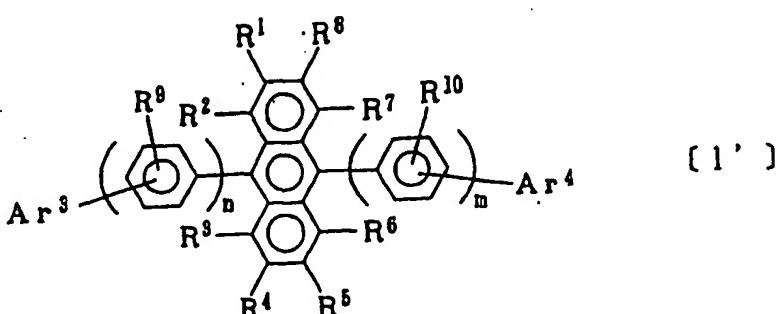
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n represents a number of 1 to 3 and m represents a number of 1 to 3; and

a case in which Ar³ and Ar⁴ both represent an alkenyl group and n and m both represent a number of 1 is excluded.

General formula [1']:



55 wherein R¹ to R¹⁰ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30

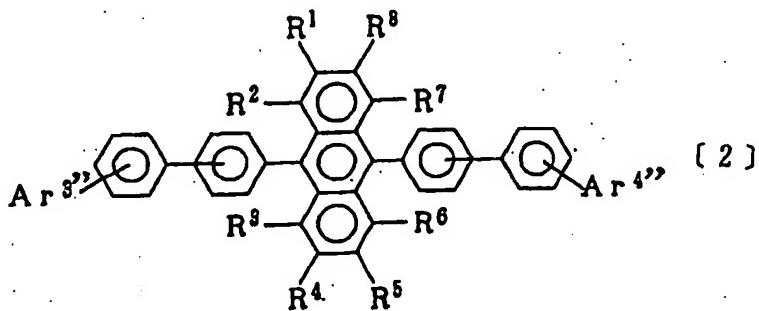
carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

[0010] Ar³ and Ar⁴ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 2 to 10 carbon atoms, the substituent in the above groups being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms; and

n represents a number of 1 to 3, m represents a number of 1 to 3 and a case in which n and m represent a same number is excluded.

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General formula [2]:



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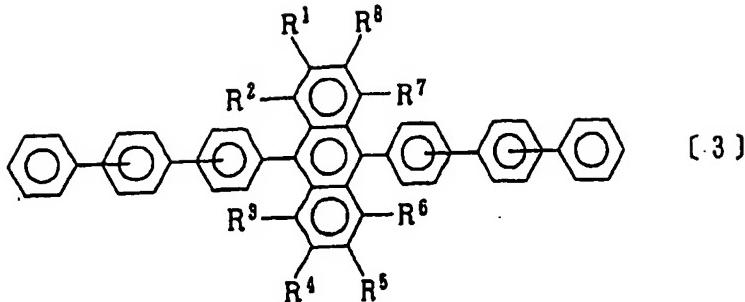
wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms; and

[0011] Ar³ and Ar⁴ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, the substituent in the above group being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 6 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms.

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General formula [3]:

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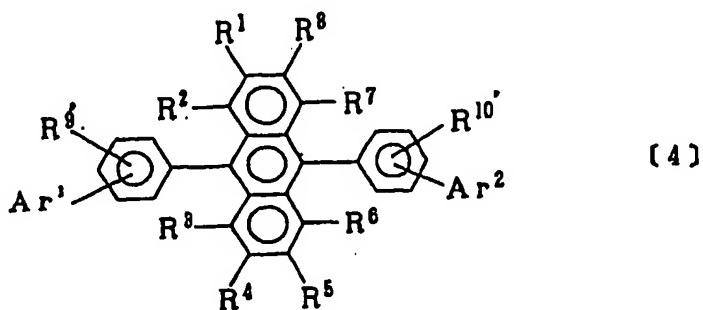


15 wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms.

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General formula [4]:



wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

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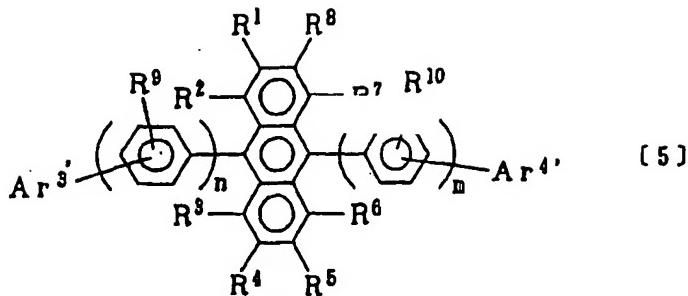
[0012] R⁹ and R¹⁰ each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; and

[0013] Ar¹ and Ar² each independently represent and a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, the substituent in the above group being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms.

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General formula [5]:



wherein R¹ to R¹⁰ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms, and at least one of R¹ to R⁸ represents the alkyl group, the alkoxy group, the aryloxy group, the alkylthio group, the arylthio group, the arylalkyl group, the monocyclic group, the condensed polycyclic group or the heterocyclic group;

[0014] Ar³ and Ar⁴ each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; and

[0015] n represents a number of 1 or 2 and m represents a number of 1 or 2.

[0016] The organic EL device of the present invention comprises a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein at least one of the layers of thin films of organic compounds comprises any of novel compounds represented by general formulae [1], [1'] and [2] to [5].

[0017] It is preferable that the light emitting layer comprises any of novel compounds represented by general formulae [1], [1'] and [2] to [5].

[0018] The light emitting layer may comprise any of novel compounds represented by general formulae [1], [1'] and [2] to [5] and a fluorescent dopant.

[0019] It is preferable that the organic EL device emits light having a peak wavelength of 460 nm or shorter.

[0020] It is preferable that the fluorescent dopant is an amine compound.

BRIEF DESCRIPTION OF THE DRAWINGS

[0021]

Figure 1 shows a spectrum of the organic EL device of Example 1 of the present invention.

THE MOST PREFERRED EMBODIMENT TO CARRY OUT THE INVENTION

[0022] The novel compound of the present invention useful for the organic EL device which emits bluish light is represented by one of the above formulae [1], [1'] and [2] to [5].

[0023] R¹ to R¹⁰ in the above general formulae [1] and [1'] each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30

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carbon atoms.

[0024] Ar³ and Ar⁴ in the above general formulae [1] and [1'] each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 2 to 10 carbon atoms. The substituent to the above groups is a substituted or unsubstituted alkyl group having 1 to 20 carbon

5 atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms.

10 [0025] In the above general formulae [1] and [1'], n represents a number of 1 to 3 and m represents a number of 1 to 3. However, the case in which Ar³ and Ar⁴ both represent an alkenyl group and n and m both represent a number of 1 is excluded in general formula [1] and the case in which n and m represent the same number is excluded in general formula [1'].

15 [0026] In the above general formula [2], R¹ to R⁸ are the same as R¹ to R⁸ defined in general formula [1].

[0027] In the above general formula [2], Ar³ⁿ and Ar⁴ⁿ each independently represent a substituted or unsubstituted aryl group having 6 to 30. The substituent in the above group is a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 6 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms.

20 [0028] In the above general formula [3], R¹ to R⁸ are the same as R¹ to R⁸ defined in general formula [1].

25 [0029] In the above general formula [4], R¹ to R⁸ are the same as R¹ to R⁸ defined in general formula [1] and R⁹ and R¹⁰ each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms.

30 [0030] In the above general formula [4], Ar¹ and Ar¹ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms. The substituent to the above group is a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms.

35 [0031] Since the groups represented by R⁸ and R¹⁰ in the compound represented by general formula [4] are alkenyl groups, the organic EL compound obtained by using this compound exhibits an enhanced fluorescent property and an improved efficiency of light emission.

[0032] In the above general formula [5], R¹ to R¹⁰ are the same as R¹ to R¹⁰ defined in general formula [1] with a proviso that at least one of R¹ to R⁸ represents the alkyl group, the alkoxy group, the aryloxy group, the alkylthio group, the arylthio group, the arylalkyl group, the monocyclic group, the condensed polycyclic group or the heterocyclic group.

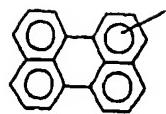
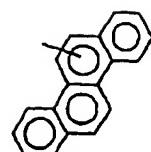
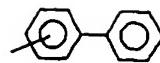
40 [0033] In the above general formula [5], Ar³ and Ar⁴ each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms.

[0034] Since at least one of R¹ to R⁸ represents the alkyl group, the alkoxy group, the aryloxy group, the alkylthio group, the arylthio group, the arylalkyl group, the monocyclic group, the condensed polycyclic group or the heterocyclic group in the above general formula [5], the organic EL compound obtained by using the compound represented by general formula [5] exhibits improved uniformity of light emission on the light emitting surface and the possibility of forming defects decreases.

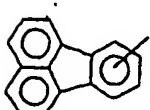
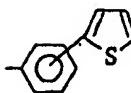
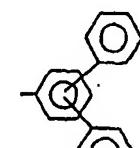
45 [0035] Examples of the groups represented by Ar¹ to Ar⁴ include the following groups:

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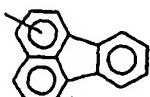
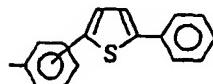
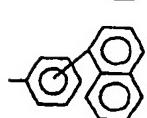
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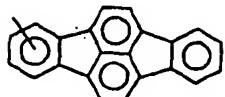
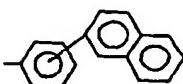
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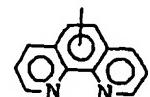
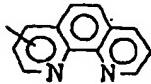
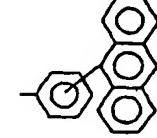
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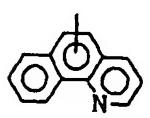
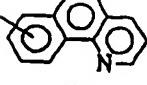
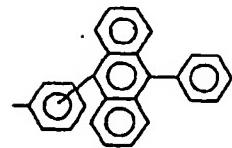
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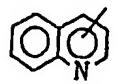
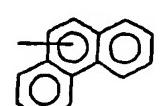
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and the above groups having alkyl groups, alkoxy groups or aryl groups as the substituents.

[0036] As described above, the organic EL device of the present invention comprises a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes and at least one of the layers of thin films of organic compounds comprises

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any of the novel compounds represented by the above general formulae [1], [1'] and [2] to [5].

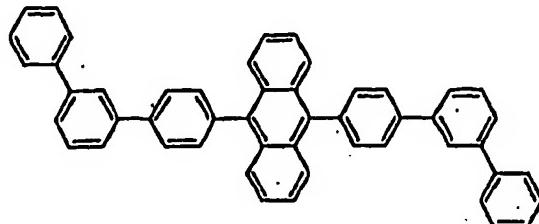
[0037] It is preferable that the layer of the thin films of organic compounds comprises 1 to 100% by mole and more preferably 10 to 98% by mole of any of the novel compounds represented by the above general formulae [1], [1'] and [2] to [5].

5 [0038] The organic EL device of the present invention emits bluish light.

[0039] Typical examples of the compounds represented by the above general formulae [1], [1'] and [2] to [5] are shown in the following as compounds (1) to (43). However, the novel compound of the present invention is not limited to the compounds shown as the examples.

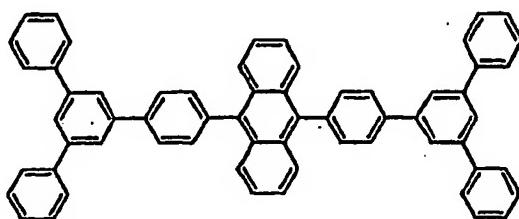
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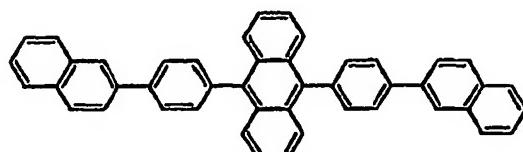
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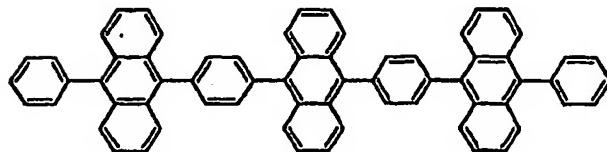
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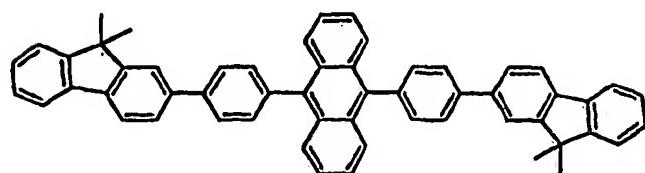
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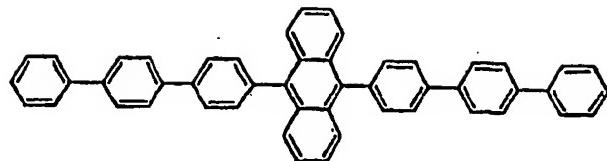
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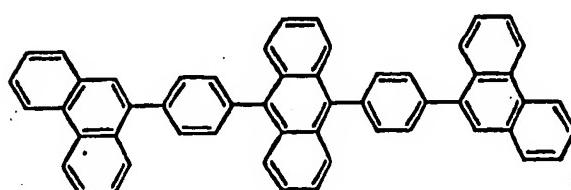


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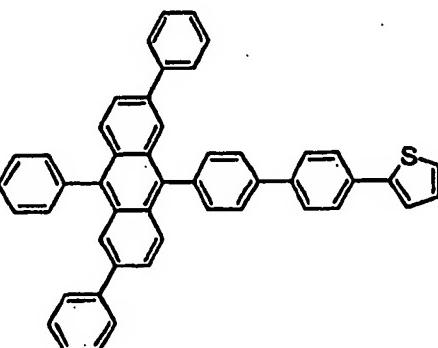


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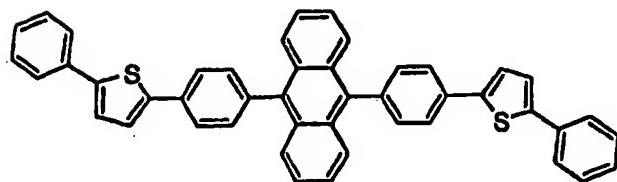


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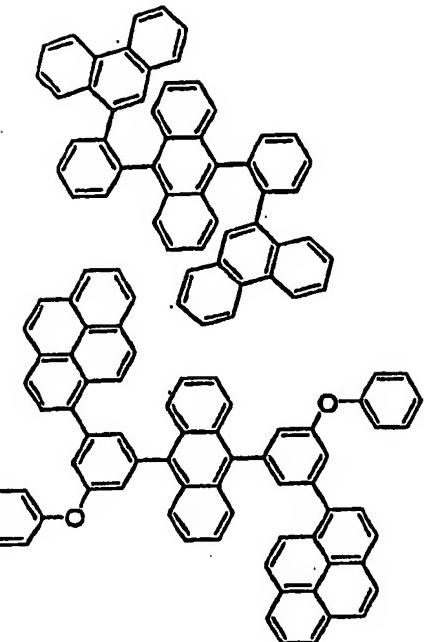
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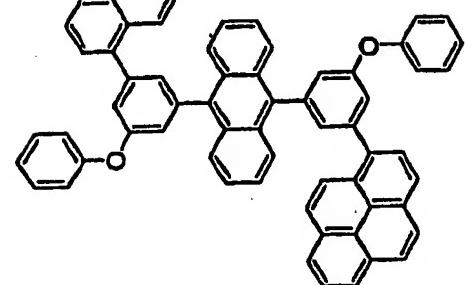
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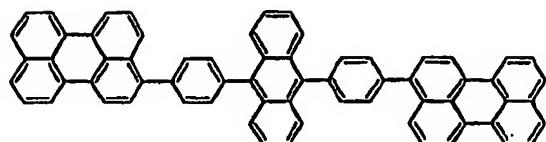
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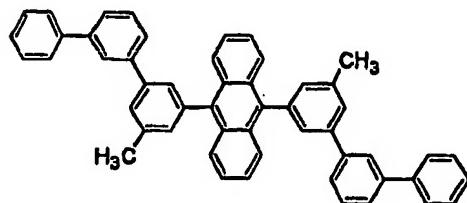
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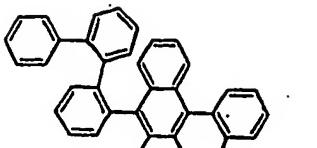


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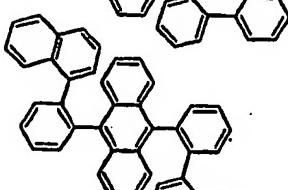
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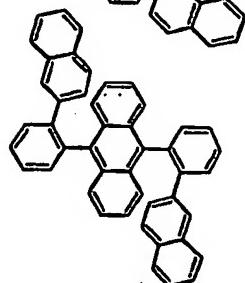
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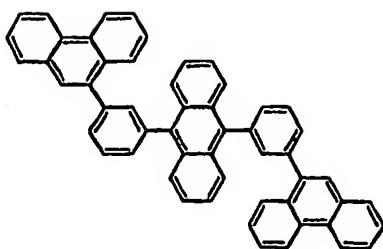


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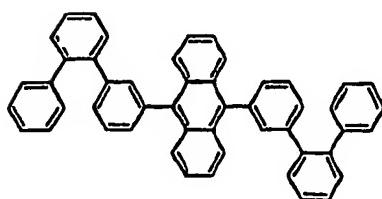
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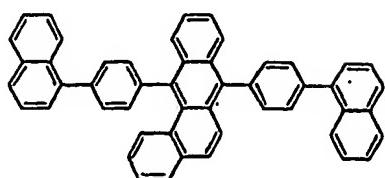
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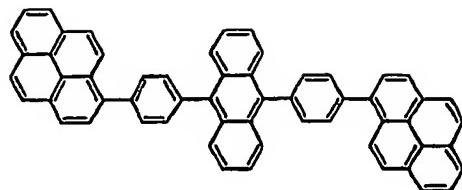
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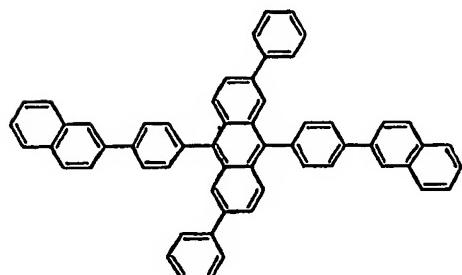
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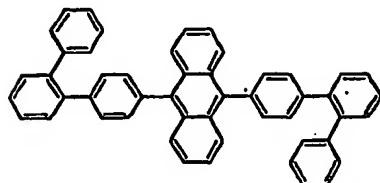


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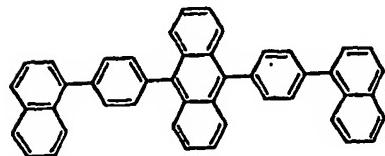
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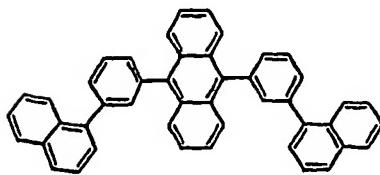
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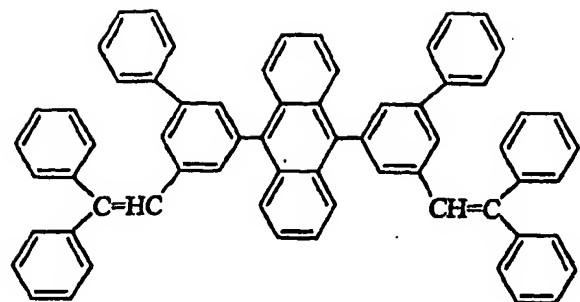
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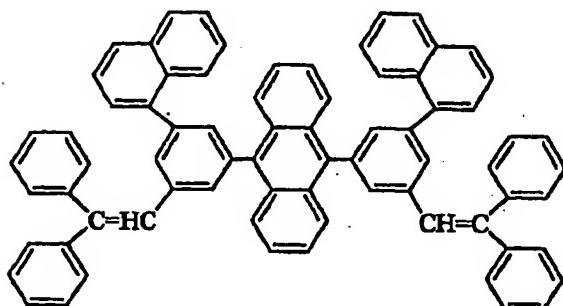


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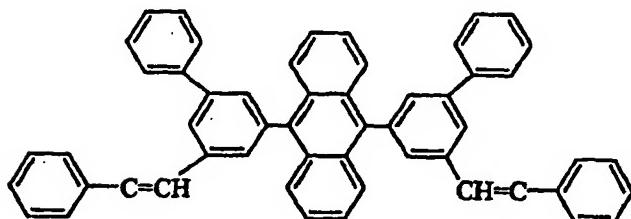


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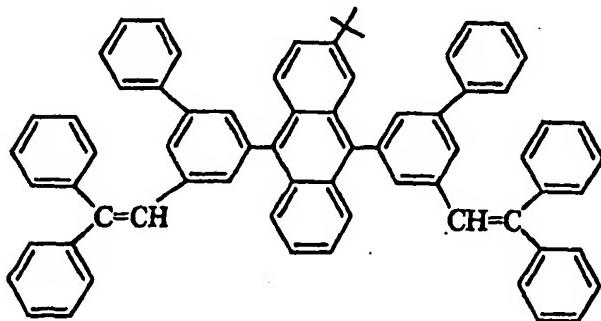
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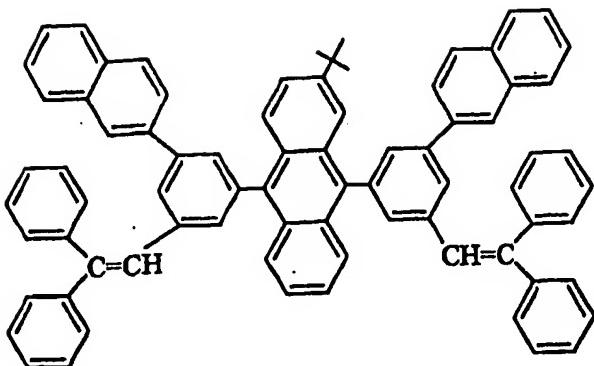
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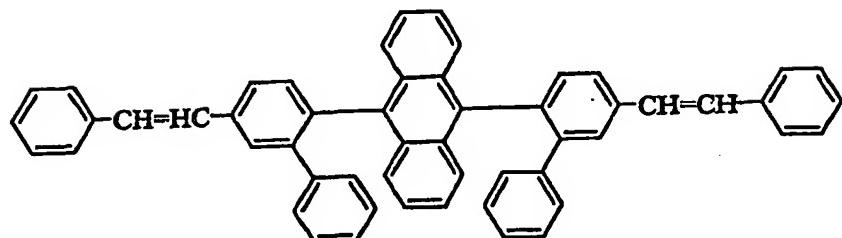
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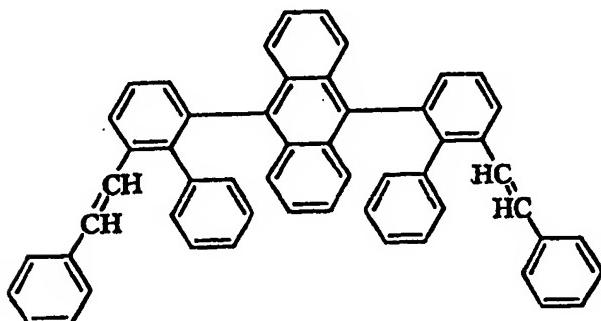
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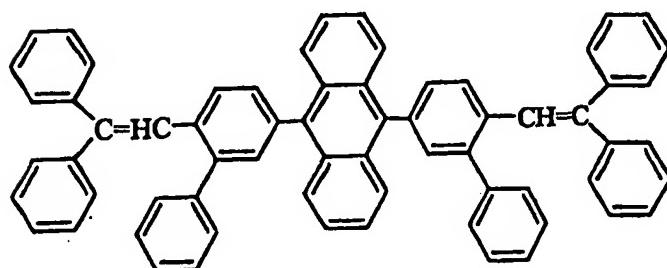
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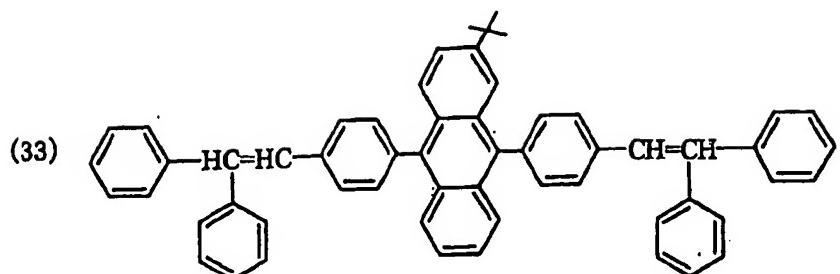
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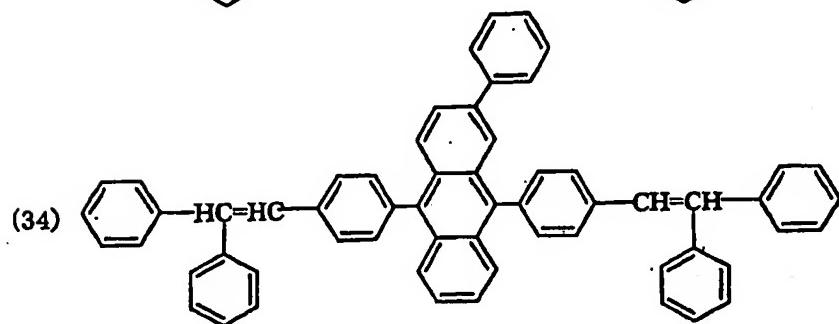
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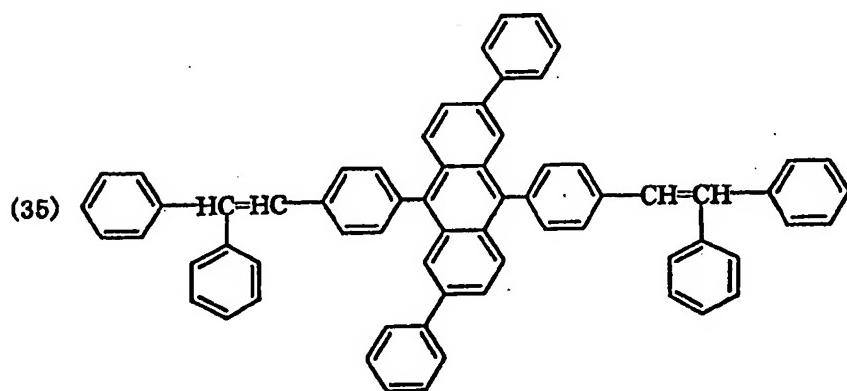


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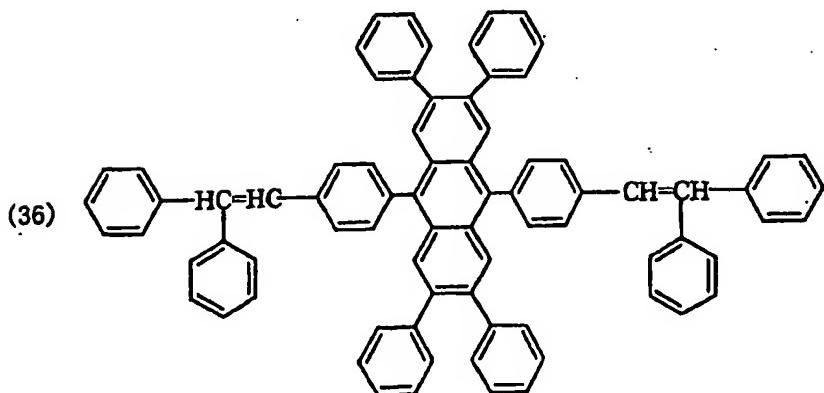
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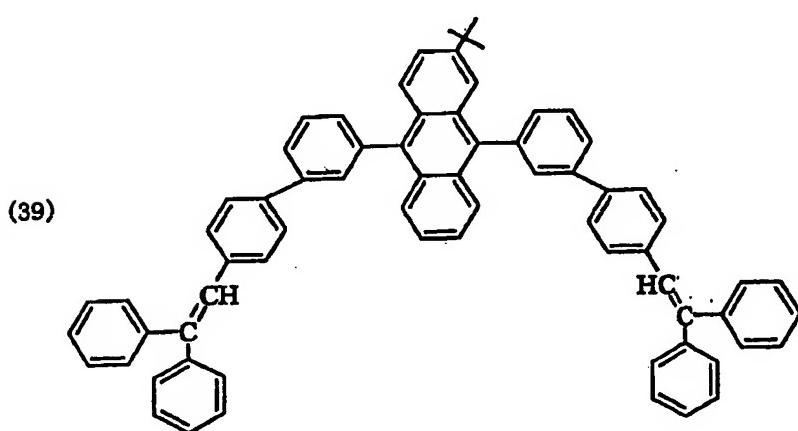
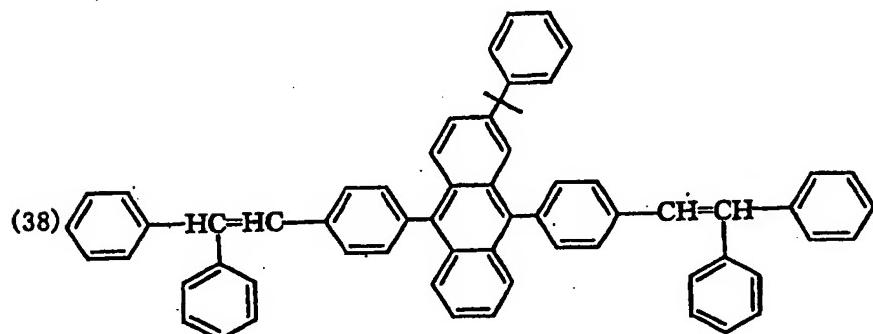
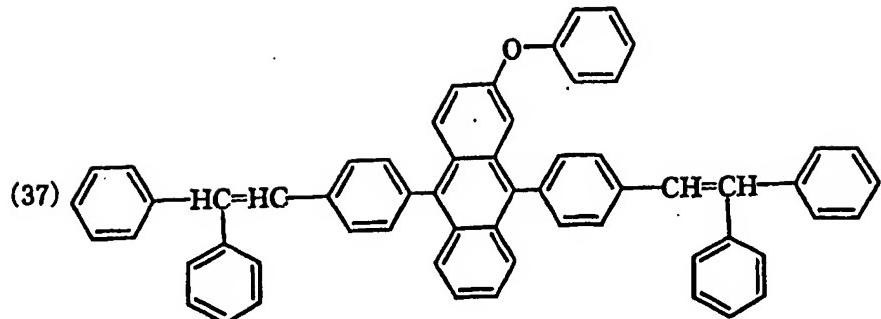
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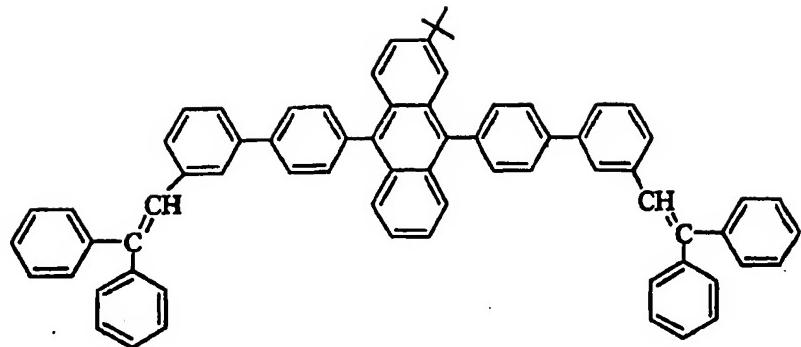
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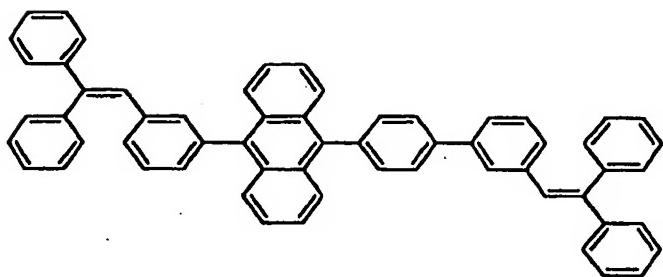
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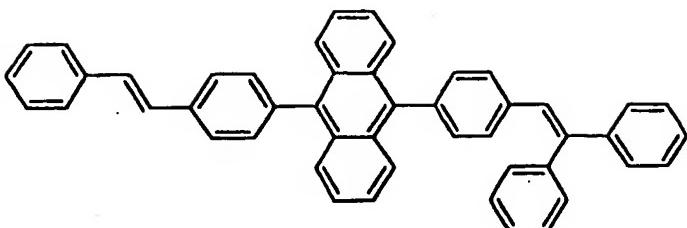


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[0040] The organic EL device of the present invention is a device in which one or a plurality of organic thin films are disposed between an anode and a cathode. When the device has a single organic layer, a light emitting layer is disposed between an anode and a cathode. The light emitting layer contains a light emitting material and may also contain a hole injecting material to transport holes injected at the anode to the light emitting material or an electron injecting material to transport electrons injected at the cathode to the light emitting material. It is preferable that the light emitting layer is formed with a light emitting material having a very high quantum efficiency of fluorescence emission and excellent ability to transfer holes and electrons and a uniform thin film is formed. The organic EL device having a multi-layer structure has a laminate structure such as: (an anode / a hole injecting layer / a light emitting layer / a cathode), (an anode / a light emitting layer / an electron injecting layer / a cathode) and (an anode / a hole injecting layer / a light emitting layer / an electron injecting layer / a cathode).

[0041] In the light emitting layer, where necessary, conventional light emitting materials, doping materials, hole injecting materials and electron injecting materials may be used in addition to any of the compounds represented by general formulae [1], [1'] and [2] to [5] used in the present invention. Deterioration in the luminance and the life caused by quenching can be prevented by the multi-layer structure of the organic EL. Where necessary, light emitting materials, other doping materials, hole injecting materials and electron injecting materials may be used in combination. By using other doping materials, the luminance and the efficiency of light emission can be improved and red light and white light can be emitted. The hole injecting layer, the light emitting layer and the electron injecting layer may each have a multi-layer structure having two or more layers. When the hole injecting layer has a multi-layer structure, the layer into which holes are injected from the electrode is referred to as the hole injecting layer and the layer which receives holes from the hole injecting layer and transports holes from the hole injecting layer to the light emitting layer is referred to as the hole transporting layer. When the electron injecting layer has a multi-layer structure, the layer into which electrons are injected from the electrode is referred to as the electron injecting layer and the layer which receives electrons from the electron injecting layer and transports electrons from the electron injecting layer to the light emitting layer is referred to as the electron transporting layer. These layers are each selected and used in accordance with factors such as the energy level, heat resistance and adhesion with the organic layers or the metal electrodes of the material.

[0042] Examples of the material which can be used in the organic layer as the light emitting material or the host material in combination with any of the compounds represented by general formulae [1], [1'] and [2] to [5] include anthracene, naphthalene, phenanthrene, pyrene, tetracene, coronene, chrysene, fluoresceine, perylene, phthaloperylene, naphthaloperylene, perynone, phthaloperynone, naphthaloperynone, diphenylbutadiene, tetraphenylbutadiene, coumarine, oxadiazole, aldazine, bisbenzoxazoline, bisstyryl, pyrazine, cyclopentadiene, metal complexes of quinoline, metal complexes of aminoquinoline, metal complexes of benzoquinoline, imines, diphenylethylene, vinylanthracene, diaminocarbazole, pyrane, thiopyrane, polymethine, merocyanine, chelates of oxinoid compounds with imidazoles, quinacridone, rubrene, stilbene derivatives and fluorescent pigments. However, the above material is not limited to the compounds described above as the examples.

[0043] As the hole injecting material, a compound which has the ability to transfer holes, exhibits an excellent effect of hole injection from the anode and an excellent effect of hole injection to the light emitting layer or the light emitting material, prevents transfer of excited components formed in the light emitting layer into the electron injecting layer or the electron injecting material and has excellent ability to form a thin film is preferable. Examples of the above compound include phthalocyanine derivatives, naphthalocyanine derivatives, porphyrin derivatives, oxazole, oxadiazole, triazole, imidazole, imidazolone, imidazolethione, pyrazoline, pyrazolone, tetrahydroimidazole, oxazole, oxadiazole, hydrazone, acylhydrazone, polyarylalkanes, stilbene, butadiene, triphenylamines of the benzidine-type, triphenylamines of the styrylamine type, triphenylamines of the diamine type, derivatives of these compounds and macromolecular com-

pounds such as polyvinylcarbazole, polysilane and conductive macromolecules. However, the above compound is not limited to the compounds described above as the examples.

[0044] Among the hole injection materials which can be used in the organic EL device of the present invention, aromatic tertiary amine derivatives and phthalocyanine derivatives are more effective.

[0045] Examples of the aromatic tertiary amine derivative include triphenylamine, tritolylamine, tolyldiphenylamine, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N,N',N'-(4-methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N,N',N'-(4-methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N'-diphenyl-N,N'-dinaphthyl-1,1'-biphenyl-4,4'-diamine, N,N'-(methylphenyl)-N,N'-(4-n-butylphenyl)phenanthrene-9,10-diamine, N,N-bis(4-di-4-tolylaminophenyl)-4-phenylcyclohexane and oligomers and polymers having a skeleton structure of these aromatic tertiary amines. However, the aromatic tertiary amine derivative is not limited to the compounds described above as the examples.

[0046] Examples of the phthalocyanine (Pc) derivative include H₂Pc, CuPc, CoPc, NiPc, ZnPc, PdPc, FePc, MnPc, ClAlPc, ClGaPc, ClInPc, ClSnPc, Cl₂SiPc, (HO)AlPc, (HO)GaPc, VOPc, TiOPc, MoOPc, GaPc-O-GaPc and corresponding derivatives of naphthalocyanine. However, the derivatives of phthalocyanine and naphthalocyanine are not limited to the compounds described above as the examples.

[0047] As the electron injecting material, a compound which has the ability to transport electrons, exhibits an excellent effect of electron injection from the cathode and an excellent effect of electron injection to the light emitting layer or the light emitting material, prevents transfer of excited components formed in the light emitting layer into the hole injecting layer and has excellent ability to form a thin film is preferable. Examples of the above compound include fluorenone, anthraquinodimethane, diphenoxquinone, thiopyrane dioxide, oxazole, oxadiazole, triazole, imidazole, perylenetetracarboxylic acid, fluorenylidenemethane, anthraquinodimethane, anthrone and derivatives of these compounds. However, the above compound is not limited to the compounds described above as the examples. The charge injecting property can be improved by adding an electron accepting material to the hole injecting material or by adding an electron donating material to the electron injecting material.

[0048] In the organic EL device of the present invention, more effective electron injecting materials are metal complex compounds and five-membered derivatives containing nitrogen.

[0049] Examples of the metal complex compound include 8-hydroxyquinolinatolithium, bis(8-hydroxyquinolinato)zinc, bis(8-hydroxyquinolinato)copper, bis(8-hydroxyquinolinato)manganese, tris(8-hydroxyquinolinato)aluminum, tris(2-methyl-8-hydroxyquinolinato)-aluminum, tris(8-hydroxyquinolinato)gallium, bis(10-hydroxybenzo-[h]quinolinato)beryllium, bis(10-hydroxybenzo[h]quinolinato)zinc, bis(2-methyl-8-quinolinato)chlorogallium, bis(2-methyl-8-quinolinato)(o-cresolato)gallium, bis(2-methyl-8-quinolinato)(1-naphtholato)aluminum and bis(2-methyl-8-quinolinato)(2-naphtholato)gallium. However, the metal complex compound is not limited to the compounds described above as the examples.

[0050] Preferable examples of the five-membered derivative containing nitrogen include derivatives of oxazoles, thiazoles, thiadiazoles and triazoles. Specific examples include 2,5-bis(1-phenyl)-1,3,4-oxazole, dimethylPOPOP, 2,5-bis(1-phenyl)-1,3,4-thiazole, 2,5-bis(1-phenyl)-1,3,4-oxadiazole, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-oxadiazole, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole, 1,4-bis[2-(5-phenyloxadiazolyl)]benzene, 1,4-bis[2-(5-phenyloxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-thiadiazole, 2,5-bis(1-naphthyl)-1,3,4-thiadiazole, 1,4-bis[2-(5-phenylthiadiazolyl)]benzene, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-triazole, 2,5-bis(1-naphthyl)-1,3,4-triazole and 1,4-bis[2-(5-phenyltriazolyl)]benzene. However, the five-membered derivative containing nitrogen is not limited to the compounds described above as the examples.

[0051] In the organic EL device of the present invention, the organic layer may contain at least one of light emitting materials, doping materials, hole injecting materials and electron injecting materials in the same layer in addition to any of the compounds represented by general formula [1], [1'] and [2] to [5]. In order to improve stability of the organic EL device of the present invention with respect to temperature, humidity and atmosphere, a protecting layer may be formed on the surface of the device or the entire device may be protected with silicon oil or a resin.

[0052] As the conductive material used for the anode of the organic EL device, a material having a work function of 4 eV or greater is suitable. Examples of such a material include carbon, aluminum, vanadium, iron, cobalt, nickel, tungsten, silver, gold, platinum, palladium, alloys of these metals, metal oxides used for ITO substrates and NESA substrates such as tin oxide and indium oxide and organic conductive resins such as polythiophene and polypyrrrol. As the conductive material used for the cathode, a material having a work function smaller than 4 eV is suitable. Examples of such a material include magnesium, calcium, tin, lead, titanium, yttrium, lithium, ruthenium, manganese, aluminum and alloys of these metals. However, the materials used for the anode and the cathode are not limited to the materials described above as the examples. Typical examples of the alloy include alloys of magnesium and silver, alloys of magnesium and indium and alloys of lithium and aluminum. However, the alloy is not limited to these alloys described as the examples. The composition of the alloy is controlled by the temperature of the source of vapor deposition, the atmosphere and the degree of vacuum and can be adjusted suitably. The anode and the cathode may have a multi-layer structure having two or more layers, where necessary.

[0053] In the organic EL device, to achieve efficient light emission, it is preferable that at least one face of the device is sufficiently transparent in the wave length region of the emitted light. It is preferable that the substrate is also trans-

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parent. The transparent electrode is disposed in accordance with vapor deposition or sputtering using the above conductive material in a manner such that the prescribed transparency is surely obtained. It is preferable that the electrode disposed on the light emitting face has a transmittance of light of 10% or greater. The substrate is not particularly limited as long as the substrate has sufficient mechanical strength and strength at high temperatures and is transparent. Glass substrates or transparent films of resins may be used. Examples of the transparent films of resins include films of polyethylene, ethylene-vinyl acetate copolymers, ethylene-vinyl alcohol copolymers, polypropylene, polystyrene, polymethyl methacrylate, polyvinyl chloride, polyvinyl alcohol, polyvinyl butyral, nylon, polyether ether ketones, polysulfones, polyether sulfones, tetrafluoroethylene-perfluoroalkyl vinyl ether copolymers, polyvinyl fluoride, tetrafluoroethylene-ethylene copolymers, tetrafluoroethylene-hexafluoropropylene copolymers, polychlorotrifluoroethylene, polyvinylidene fluoride, polyesters, polycarbonates, polyurethanes, polyether imides, polyimides and polypropylene.

[0054] Each layer of the organic EL device of the present invention can be formed suitably in accordance with a dry process of film formation such as vacuum vapor deposition, sputtering, plasma plating and ion plating or a wet process of film formation such as spin coating, dipping and flow coating. The thickness of the film is not particularly limited. However, it is necessary that the thickness be set at a suitable value. When the thickness is greater than the suitable value, a high voltage must be applied to obtain a prescribed output of light and the efficiency decreases. When the thickness is smaller than the suitable value, pin holes are formed and a sufficient luminance cannot be obtained even when the electric field is applied. In general, the suitable range of the thickness is 5 nm to 10 μm . A thickness in the range of 10 nm to 0.2 μm is preferable.

[0055] When the device is produced in accordance with a wet process, materials forming each layer are dissolved or dispersed in a suitable solvent such as ethanol, chloroform, tetrahydrofuran and dioxane and a film is formed from the solution or the suspension. The solvent is not particularly limited. In any organic thin layer, suitable resins and additives may be used to improve the property to form a film and to prevent formation of pin holes. Examples of the resin which can be used include insulating resins such as polystyrene, polycarbonates, polyarylates, polyesters, polyamides, polyurethanes, polysulfones, polymethyl methacrylate, polymethyl acrylate and cellulose; copolymers derived from these resins; photoconductive resins such as poly-N-vinylcarbazole and polysilane; and conductive resins such as polythiophene and polypyrrol. Examples of the additive include antioxidants, ultraviolet light absorbents and plasticizers.

[0056] As described above, when the compound of the present invention is used for the organic layer of the organic EL device, the organic EL device exhibiting excellent efficiency of light emission and heat resistance, having a long life and emitting bluish light having excellent purity of color can be obtained.

[0057] The organic EL device of the present invention can be used for a planar light emitting member such as a flat panel display of wall televisions, a back light for copiers, printers and liquid crystal displays, a light source of instruments, display panels and a marker light.

[0058] The present invention will be described more specifically with reference to Synthesis Examples and Examples in the following.

Synthesis Example 1 (Compound 14)

(1) Synthesis of 2,2'-dibromobiphenyl

[0059] Under an atmosphere of argon, 1,2-bromobenzene (25 g, 0.11 mole) was dissolved into anhydrous THF (240 ml) and the resultant solution was cooled at -67°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 35 ml, 53 mmole, 0.5 eq) was slowly added dropwise over 5 minutes. The resultant mixture was stirred at -67°C for 1 hour and then at the room temperature for 3 hours. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (100 ml) was added. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried over anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a brown liquid (about 19 g) was obtained. The obtained liquid was purified by the column chromatography (silica gel/hexane) and white needle crystals (9.5 g, 57%) were obtained.

$^1\text{H-NMR}$ (CDCl_3 , TMS) δ : 7.2 to 7.4 (6H, m), 7.6 to 7.7 (2H, m)

(2) Synthesis of 2-phenyl-2'-bromobiphenyl

[0060] Under an atmosphere of argon, 2,2'-dibromobiphenyl (9.5 g, 30 mmole), phenylboronic acid (3.7 g, 30 mmole) and tetrakis(triphenylphosphine)palladium(0) (1.0 g, 0.87 mmole, 3% Pd) were dissolved into toluene (75 ml). To the obtained solution, an aqueous solution of sodium carbonate (9.7 g, 92 mmole, 3 eq/46 ml) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried over anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a dark yellow oil was obtained. The obtained oil was purified

by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+5% dichloromethane, successively) and a colorless oil (6.5 g, 70%) was obtained.

$^1\text{H-NMR}$ (CDCl_3 , TMS) δ : 6.6 to 6.7 (1H, m), 7.0 to 7.6 (12H, m)

5 (3) Synthesis of 9,10-bis(2-(2-phenylphenyl)phenyl)-9,10-dihydroanthracene

[0061] Under an atmosphere of argon, 2-phenyl-2'-bromobiphenyl (6.5 g, 21 mmole, 2.5 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous tetrahydrofuran (THF) (25 ml) and the resultant solution was cooled at -30°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 15 ml, 23 mmole, 1.1 eq) was added and the resultant solution was stirred at -20°C for 1 hour. To the obtained solution, anthraquinone (1.7 g, 8.2 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration, washed with water, methanol and acetone and a white solid (2.9 g, 53%) was obtained.

10 15 $^1\text{H-NMR}$ (CDCl_3 , TMS) δ : 0.47 (2H, s), 5.7 to 5.8 (2H, m), 6.3 to 7.4 (30H, m), 8.3 to 8.4 (2H, m)

(4) Synthesis of 9,10-bis(2-(2-phenylphenyl)phenyl)anthracene (Compound 14)

[0062] 9,10-bis(2-(2-Phenylphenyl)phenyl)-9,10-dihydroanthracene (2.9 g, 4.3 mmole) was suspended in acetic acid (45 ml). To the resultant suspension, a 57% hydroiodic acid (6 ml, 45 mmole, 10 eq) was added and the obtained mixture was stirred at 100°C for 6 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (30 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a light yellow solid (2.4 g, 88%) was obtained.

20 25 $^1\text{H-NMR}$ (CDCl_3 , TMS) δ : 6.7 to 7.5 (34H, m), all-H

Synthesis Example 2 (Compound 15)

(1) Synthesis of 1-(2-bromophenyl)naphthalene

[0063] Under an atmosphere of argon, 2-bromoiodobenzene (7.0 g, 25 mmole), naphthaleneboronic acid (4.0 g, 23 mmole) and tetrakis(triphenylphosphine)palladium(0) (0.5 g, 0.43 mmole, 1.7% Pd) were dissolved into toluene (50 ml). To the obtained solution, an aqueous solution of sodium carbonate (7.3 g, 69 mmole, 3 eq/35 ml) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+10% dichloromethane, successively) and white needle crystals (5.4 g, 83%) were obtained.

30 35 $^1\text{H-NMR}$ (CDCl_3 , TMS) δ : 7.3 to 7.8 (9H, m), 7.90 (2H, dd, $J=8$ Hz, 2 Hz)

(2) Synthesis of 9,10-bis(2-(1-naphthyl)phenyl)-9,10-dihydroanthracene

[0064] Under an atmosphere of argon, 1-(2-bromophenyl)naphthalene (5.4 g, 19 mmole, 2.8 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 14 ml, 21 mmole, 1.1 eq) was added and the resultant solution was stirred at -20°C for 1 hour. To the obtained solution, anthraquinone (1.4 g, 6.7 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 3 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration, washed with water, methanol and acetone and a white solid (3.5 g, 85%) was obtained.

40 50 $^1\text{H-NMR}$ (CDCl_3 , TMS) δ : -0.20 (2H, s), 5.76 (2H, dd, $J=13$ Hz, 7 Hz), 6.2 to 7.7 (26H, m), 8.43 (2H, d, $J=8$ Hz)

(3) Synthesis of 9,10-bis(2-(1-naphthyl)phenyl)anthracene (Compound 15)

[0065] 9,10-bis(2-(1-Naphthyl)phenyl)-9,10-dihydroanthracene (3.5 g, 5.7 mmole) was suspended in acetic acid (80 ml). To the resultant suspension, a 57% hydroiodic acid (15 ml, 0.11 mole, 20 eq) was added and the obtained mixture was stirred at 100°C for 7 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (30 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (3.2 g, 96%) was obtained.

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¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (30H, m), all-H

Synthesis Example 3 (Compound 17)

5 (1) Synthesis of 9-phenanthreneboronic acid

[0066] Under an atmosphere of argon, 9-bromophenanthrene (15 g, 58 mmole) was dissolved into anhydrous ether (150 ml) and the resultant solution was cooled at -35°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 43 ml, 65 mmole) was added dropwise and the resultant mixture was stirred at -20°C for 1 hour. After the reaction mixture was cooled at -67°C, a solution (30 ml) of triisopropoxyborane (37 ml, 0.16 mole, 2.8 eq) in anhydrous ether was added the resultant mixture was stirred at -65°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a 10% hydrochloric acid (150 ml) was added. After the resultant mixture was stirred at the room temperature for 1 hour, the formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. The solid obtained after removing the solvent by distillation was washed with hexane and a white solid (10 g, 78%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.6 to 7.9 (5H, m), 8.17 (1H, s), 8.5 to 8.8 (3H, m)

20 (2) Synthesis of 9-(3-bromophenyl)phenanthrene

[0067] Under an atmosphere of argon, 3-bromoiodobenzene (7.0 g, 25 mmole), phenanthreneboronic acid (5.0 g, 23 mmole) and tetrakis-(triphenylphosphine)palladium(0) (0.5 g, 0.43 mmole, 1.7% Pd) were dissolved into toluene (100 ml). To the obtained solution, an aqueous solution of sodium carbonate (7.3 g, 69 mmole, 3 eq/35 ml) was added and the resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+5% dichloromethane, successively) and white needle crystals (6.5 g, 85%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.3 to 8.7 (11H, m), 8.76 (2H, d, J=7 Hz)

30 (3) Synthesis of 9,10-bis(3-(9-phenanthryl)phenyl)-9,10-dihydroanthracene

[0068] Under an atmosphere of argon, 1-(3-bromophenyl)naphthalene (6.5 g, 20 mmole, 2.8 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -25°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 15 ml, 23 mmole, 1.1 eq) was added and the resultant solution was stirred at -20°C for 1 hour. To the obtained mixture, anthraquinone (1.5 g, 7.2 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 3 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (5.9 g, quant.) was obtained.

¹H-NMR (CDCl₃, TMS) δ: -0.16 (2H, s), 6.06 (2H, s), 6.4 to 7.0 (12H, m), 7.1 to 7.7 (12H, m), 8.20 (2H, dd, J=8 Hz, 2 Hz), 8.4 to 8.6 (6H, m)

45 (4) Synthesis of 9,10-bis(3-(9-phenanthryl)phenyl)anthracene (Compound 17)

[0069] 9,10-bis(3-(9-Phenanthryl)phenyl)-9,10-dihydroanthracene (5.2 g, 7.3 mmole) was suspended in acetic acid (120 ml). To the resultant suspension, a 57% hydroiodic acid (10 ml, 77 mmole, 10 eq) was added and the obtained mixture was stirred at 100°C for 6 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (40 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (5.0 g, quant.) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.0 to 8.5 (34H, m), all-H

Synthesis Example 4 (Compound 20)

55 (1) Synthesis of 1-(4-bromophenyl)pyrene

[0070] Under an atmosphere of argon, 4-bromoiodobenzene (7.0 g, 25 mmole), 1-pyreneboronic acid (5.7 g, 23 mmole) and tetrakis(triphenylphosphine)palladium(0) (0.5 g, 0.43 mmole, 1.7% Pd) were dissolved into toluene (50

ml). To the obtained solution, an aqueous solution of sodium carbonate (7.3 g, 69 mmole, 3 eq/35 ml) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+10% dichloromethane, successively) and white needle crystals (6.6 g, 80%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.3 to 7.8 (11H, m), 7.90 (2H, d, J=8 Hz)

(2) Synthesis of 9,10-bis(4-(1-pyrenyl)phenyl)-9,10-dihydroanthracene

[0071] Under an atmosphere of argon, 1-(2-bromophenyl)pyrene (6.6 g, 18 mmole, 2.8 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 14 ml, 21 mmole, 1.1 eq) was added and the obtained solution was stirred at -20°C for 1 hour. To the obtained mixture, anthraquinone (1.4 g, 6.7 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 3 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (4.5 g, 88%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: -0.20 (2H, s), 5.76 (2H, dd, J=13 Hz, 7 Hz), 6.2 to 7.7 (30H, m), 8.43 (2H, d, J=8 Hz)

(3) Synthesis of 9,10-bis(4-(1-pyrenyl)phenyl)anthracene (Compound 20)

[0072] 9,10-bis(4-(1-Pyrenyl)phenyl)-9,10-dihydroanthracene (4.5 g, 5.9 mmole) was suspended in acetic acid (80 ml). To the resultant suspension, a 57% hydroiodic acid (15 ml, 0.11 mole, 20 eq) was added and the obtained mixture was stirred at 100°C for 7 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (30 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (3.9 g, 90%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (30H, m), all-H

Synthesis Example 5 (Compound 22)

(1) Synthesis of 2-biphenylboronic acid

[0073] Under an atmosphere of argon, 2-bromobiphenyl (20 g, 86 mmole) was dissolved into anhydrous ether (200 ml) and the resultant solution was cooled at -35°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter; 63 ml, 95 mmole) was added dropwise and the resultant mixture was stirred at -20°C for 1 hour. After the reaction mixture was cooled at -67°C, a solution (50 ml) of triisopropoxyborane (50 ml, 0.22 mole, 2.5 eq) in anhydrous ether was added and the resultant mixture was stirred at -65°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a 10% hydrochloric acid (200 ml) was added. After the resultant mixture was stirred at the room temperature for 1 hour, the formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. The solid obtained after removing the solvent by distillation was washed with hexane and a white solid (11 g, 62%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.74(1H, d, J=7 Hz), 7.1 to 7.4 (8H, m)

(2) Synthesis of 2-(4-bromophenyl)biphenyl

[0074] Under an atmosphere of argon, 2-bromoiodobenzene (7.9 g, 25 mmole), biphenylboronic acid (5.0 g, 25 mmole) and tetrakis(triphenylphosphine)palladium(0) (0.5 g, 0.43 mmole, 1.7% Pd) were dissolved into toluene (60 ml). To the obtained solution, an aqueous solution of sodium carbonate (8.0 g, 75 mmole, 3 eq/40 ml) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+5% dichloromethane, successively) and white needle crystals (6.8 g, 88%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.3 to 8.7 (11H, m), 8.76 (2H, d, J=7 Hz)

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(3) Synthesis of 9,10-bis(4-(2-phenylphenyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene

[0075] Under an atmosphere of argon, 2-(4-bromophenyl)biphenyl (6.8 g, 22 mmole, 2.5 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -30°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 15 ml, 23 mmole, 1.1 eq) was added and the resultant solution was stirred at -20°C for 1 hour. To the obtained mixture, anthraquinone (1.7 g, 8.2 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (4.9 g, 89%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 5.7 to 5.8 (2H, m), 6.3 to 7.4 (30H, m), 8.3 to 8.4 (2H, m)

(4) Synthesis of 9,10-bis(4-(2-phenylphenyl)phenyl)anthracene (Compound 22)

[0076] 9,10-bis(4-(2-Phenylphenyl)phenyl)-9, 10-dihydroxy-9,10-dihydroanthracene (4.9 g, 7.3 mmole) was suspended in acetic acid (70 ml). To the resultant suspension, a 57% hydroiodic acid (10 ml, 77 mmole, 10 eq) was added and the obtained mixture was stirred at 100°C for 6 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (50 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a light yellow solid (4.6 g, 88%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (34H, m), all-H

Synthesis Example 6 (Synthesis of Compound 1)

(1) Synthesis of 9,10-bis(4-(3-phenylphenyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene

[0077] Under an atmosphere of argon, 3-(4-bromophenyl)biphenyl (6.8 g, 22 mmole, 2.5 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -30°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 15 ml, 23 mmole, 1.1 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained solution, anthraquinone (1.7 g, 8.2 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (5.0 g, 91%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 5.7 to 5.8 (2H, m), 6.3 to 7.4 (30H, m), 8.3 to 8.4 (2H, m)

(2) Synthesis of 9,10-bis(4-(3-phenylphenyl)phenyl)anthracene (Compound 1)

[0078] 9,10-bis(4-(3-Phenylphenyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene (4.9 g, 7.3 mmole) was suspended in acetic acid (70 ml). To the resultant suspension, a 57% hydroiodic acid (10 ml, 77 mmole, 10 eq) was added and the obtained mixture was stirred at 100°C for 6 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (50 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a light yellow solid (4.1 g, 79%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (34H, m), all-H

Synthesis Example 7 (Synthesis of Compound 2)

(1) Synthesis of 9,10-bis(4-(3,5-diphenylphenyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene

[0079] Under an atmosphere of argon, 1,3-diphenyl-5-(4-bromophenyl)-benzene (8.5 g, 22 mmole, 2.5 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -30°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 15 ml, 23 mmole, 1.1 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained solution, anthraquinone (1.7 g, 8.2 mmole) was added and the obtained mixture was stirred at -20°C for 1 hour and at the room temperature for 2 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (16 g, 90%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 5.7 to 5.8 (2H, m), 6.3 to 7.4 (30H, m), 8.3 to 8.4 (2H, m)

(2) Synthesis of 9,10-bis(4-(3,5-diphenylphenyl)phenyl)anthracene (Compound 2)

[0080] 9,10-bis(4-(3,5-Diphenylphenyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene (6.0 g, 7.3 mmole) was suspended in acetic acid (70 ml). To the resultant suspension, a 57% hydroiodic acid (10 ml, 77 mmole, 10 eq) was added and the obtained mixture was stirred at 100°C for 6 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (50 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a light yellow solid (5.3 g, 93%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (34H, m), all-H

10 Synthesis Example 8 (Synthesis of Compound 3)

(1) Synthesis of 9,10-bis(4-(2-naphthyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene

[0081] Under an atmosphere of argon, 2-(4-bromophenyl)naphthalene (5.4 g, 19 mmole, 2.8 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.50 mole/liter, 14 ml, 21 mmole, 1.1 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the resultant solution, anthraquinone (1.4 g, 6.7 mmole) was added and the obtained mixture was stirred at -20°C for 1 hour and at the room temperature for 3 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (20 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (3.7 g, 91%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: -0.20 (2H, s), 5.76 (2H, dd, J=13 Hz, 7 Hz), 6.2 to 7.7 (26H, m), 8.43 (2H, d, J=8 Hz)

25 (2) Synthesis of 9,10-bis(4-(2-naphthyl)phenyl)anthracene (Compound 3)

[0082] 9,10-bis(4-(2-Naphthyl)phenyl)-9,10-dihydroxy-9,10-dihydroanthracene (3.5 g, 5.7 mmole) was suspended in acetic acid (80 ml). To the resultant suspension, a 57% hydroiodic acid (15 ml, 0.11 mole, 20 eq) was added and the obtained mixture was stirred at 100°C for 7 hours. To the reaction mixture, a 50% aqueous solution of hypophosphorous acid (30 ml) was added. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (3.3 g, 98%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.7 to 7.5 (30H, m), all-H

Synthesis Example 9 (Compound 25)

35 (1) Synthesis of 1-(2,2-diphenylvinyl)-3,5-dibromobenzene

[0083] Under an atmosphere of argon, 3,5-dibromobenzaldehyde (12.1 g, 46 mmole) and diethyl diphenylmethylphosphonate (15 g, 49 mmole, 1.1 eq) were dissolved into dimethyl sulfoxide (DMSO) (60 ml). To the resultant solution, potassium t-butoxide (6.2 g, 55 mmole, 1.2 eq) was added in small portions and the obtained solution was stirred at the room temperature for 9 hours and left standing for one night. After water (60 ml) was added, the reaction mixture was subjected to extraction with ethyl acetate (250 ml). The obtained organic layer was washed with water (100 ml) and a saturated aqueous solution of sodium chloride (50 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a deep brown oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane and hexane+3% dichloromethane, successively) and a white solid (14.0 g, 74%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.80 (1H, s), 7.03 (2H, d, J=2 Hz), 7.3 to 7.4 (11H, m)

(2) Synthesis of 1-(2,2-diphenylvinyl)-3-phenyl-5-bromobenzene

[0084] Under an atmosphere of argon, 1-(2,2-diphenylvinyl)-3,5-dibromobenzene (7.0 g, 17 mmole), phenylboronic acid (2.1 g, 17 mmole) and tetrakis(triphenylphosphine)palladium(0) (0.4 g, 0.35 mmole, 2% Pd) were dissolved into toluene (40 ml). To the obtained solution, a 2 M aqueous solution of sodium carbonate (25 ml, 51 mmole, 3 eq) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+3% dichloromethane and hexane+10% dichloromethane, successively) and white needle crystals (3.9 g, 56%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.94 (1H, s), 7.1 to 7.5 (18H, m)

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(3) Synthesis of 9,10-bis(3-(2,2-diphenylvinyl)phenyl-5-phenyl)-9,10-dihydroanthracene

[0085] Under an atmosphere of argon, 1-(2,2-diphenylvinyl)-3-phenyl-5-bromobenzene (3.9 g, 9.5 mmole, 2.7 eq) was dissolved into a mixed solvent composed of anhydrous toluene (20 ml) and anhydrous THF (20 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.60 mole/liter, 6 ml, 9.6 mmole, 1.0 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained solution, anthraquinone (0.7 g, 3.4 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 7 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (50 ml) was added and the reaction was deactivated. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+50% dichloromethane, dichloromethane and dichloromethane+3% methanol, successively) and a light yellow amorphous solid (2.0 g, 67%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 2.56 (2H, s), 6.5 to 6.6 (4H, m), 6.8 to 7.4 (34H, m), 7.41 (4H, dd, J=6 Hz, 3 Hz), 7.71 (4H, dd, J=6 Hz, 3 Hz)

(4) Synthesis of 9,10-bis(3-(2,2-diphenylvinyl)phenyl-5-phenyl)-anthracene (Compound 25)

[0086] 9,10-bis(3-(2,2-Diphenylvinyl)phenyl-5-phenyl)-9, 10-dihydroxy-9,10-dihydroanthracene (2.0 g, 2.3 mmole) was dissolved into acetic acid (25 ml). To the resultant solution, potassium iodide (1.5 g, 90 mmole, 4 eq) was added and the obtained solution was stirred for 3 hours. To the reaction mixture, a 50% aqueous solution of phosphinic acid was added and the reaction was deactivated. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (1.4 g, 73%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.2 to 7.4 (42H, m), 7.65 (4H, dd, J=7 Hz, 3 Hz)

[0087] The solid obtained above (1.4 g) was purified by sublimation at 380°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (0.8 g) was obtained.

FDMS: calc. for C₆₆H₄₆=838, found m/z=838 (M⁺, 4)

λ_{max}, 398, 358, 306 nm (PhMe)

Fmax, 416, 435 nm (PhMe, λ_{εx}=395 nm)

Eg=3.00 eV

I_p=5.87 eV(51 Y/eV, 100 nW)

T_g=130°C

Synthesis Example 10 (Compound 26)

(1) Synthesis of 1-(2,2-diphenylvinyl)-3-(1-naphthyl)-5-bromobenzene

[0088] Under an atmosphere of argon, 1-(2,2-diphenylvinyl)-3,5-dibromobenzene (8.3 g, 20 mmole), 1-naphthaleneboronic acid (3.4 g, 20 mmole) and tetrakis(triphenylphosphine)palladium(0) (0.46 g, 0.4 mmole, 2% Pd) were dissolved into toluene (50 ml). To the obtained solution, a 2 M aqueous solution of sodium carbonate (30 ml, 60 mmole, 3 eq) was added. The resultant solution was heated under refluxing for 10 hours and left standing for one night. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane, hexane+10% dichloromethane, hexane+10% dichloromethane, successively) and a white glass solid (5.5 g, 60%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 6.96 (1H, s), 7.1 to 7.6 (18H, m), 7.8 to 7.9 (2H, m)

(2) Synthesis of 9,10-bis(3-(2,2-diphenylvinyl)phenyl-5-(1-naphthyl))-9, 10-dihydroxy-9,10-dihydroanthracene

[0089] Under an atmosphere of argon, 1-(2,2-diphenylvinyl)-3-(1-naphthyl)-5-bromobenzene (5.5 g, 12 mmole, 2.7 eq) was dissolved into a mixed solvent composed of anhydrous toluene (30 ml) and anhydrous THF (30 ml) and the resultant solution was cooled at -30°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.60 mole/liter, 8 ml, 13 mmole, 1.0 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained mixture, anthraquinone (0.9 g, 4.4 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 7 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (50 ml) was added and the reaction was deactivated. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained.

The obtained oil was purified by the column chromatography (silica gel/hexane+50% dichloromethane and dichloromethane, successively) and a white amorphous solid (2.7 g, 63%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 2.56 (2H, s), 6.5 to 6.8 (6H, m), 6.9 to 7.5 (36H, m), 7.6 - 7.8 (8H, m)

5 (3) Synthesis of 9,10-bis(3-(2,2-diphenylvinyl)phenyl-5-(1-naphthyl))-anthracene (Compound 26)

[0090] 9,10-bis(3-(2,2-Diphenylvinyl)phenyl-5-(1-naphthyl))-9,10-dihydroanthracene (2.7 g, 2.8 mmole) was dissolved into acetic acid (30 ml). To the resultant solution, potassium iodide (1.8 g, 11 mmole, 4 eq) was added and the obtained solution was stirred for 3 hours. To the reaction mixture, a 50% aqueous solution of phosphinic acid (40 ml) was added and the reaction was deactivated. The formed solid was separated by filtration and washed with water, methanol and acetone and a white solid (2.0 g, 78%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.2 to 7.5 (40H, m), 7.7 to 7.9 (10H, m)

[0091] The solid obtained above (2.0 g) was purified by sublimation at 400°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (1.2 g) was obtained.

15 FDMS: calc. for C₇₄H₅₀=938, found m/z=938 (M⁺, 100), 469 (M²⁺, 6)

λmax, 398, 377, 358 nm (PhMe)

Fmax, 418, 436 nm (PhMe, λex=395 nm)

Eg=3.00 eV

Ip=5.86 eV(34 Y/eV, 100 nW)

Tg=132°C

Synthesis Example 11 (Compound 33)

(1) Synthesis of 2-t-butyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-9,10-dihydroanthracene

[0092] Under an atmosphere of argon, 4-(2,2-diphenylvinyl)bromobenzene (5.0 g, 15 mmole, 2.6 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.60 mole/liter, 10 ml, 16 mmole, 1.1 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained mixture, 2-(t-butyl)anthraquinone (1.5 g, 5.7 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 7 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (50 ml) was added and the reaction was deactivated. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with anhydrous magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane+50% dichloromethane, dichloromethane and dichloromethane+1% methanol, successively) and a white amorphous solid (3.3 g, 75%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 1.29 (9H, s), 2.65 (1H, s), 2.71 (1H, s), 6.68 (9H, s), 6.84 (2H, s), 7.1 to 7.4 (23H, m), 7.5 - 7.7 (4H, m)

(2) Synthesis of 3-t-butyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-anthracene (Compound 33)

[0093] 2-t-Butyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-9,10-dihydroanthracene (3.3 g, 4.3 mmole) was dissolved into acetic acid (30 ml). To the resultant solution, potassium iodide (1.9 g, 11 mmole, 2.7 eq) and sodium phosphinate monohydrate (0.6 g, 5.7 mmole) were added and the obtained mixture was stirred for 2 hours. The reaction mixture was filtered and washed with water, methanol and acetone and a light yellow solid (2.8 g, 88%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 1.28 (9H, s), 7.14 (2H, s), 7.2 to 7.5 (30H, m), 7.6 to 7.7 (5H, m)

[0094] The solid obtained above (2.8 g) was purified by sublimation at 360°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (2.2 g) was obtained.

FDMS: calc. for C₅₈H₄₆=742, found m/z=742 (M⁺, 100), 371 (M²⁺, 4)

λmax, 397, 379, 360, 310 nm (PhMe)

Fmax, 450 nm (PhMe, λex=397 nm)

Eg=2.92 eV

Ip=5.71 eV(39 Y/eV, 100 nW)

Tg=105°C

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Synthesis Example 12 (Compound 34)

(1) Synthesis of 2-phenylanthraquinone

[0095] Under an atmosphere of argon, 2-chloroanthraquinone (5.0 g, 21 mmole), phenylboronic acid (2.8 g, 23 mmole, 1.1 eq), tris(dibenzylideneacetone)dipalladium(0) (0.2 g, 0.22 mmole, 2% Pd) and potassium fluoride (4.4 g, 76 mmole, 3.3 eq) were suspended in anhydrous dioxane (30 ml). To the obtained suspension, a toluene solution of tri-t-butylphosphine (66%, 0.13 ml, 0.42 mmole, 1 eq) was added and the resultant mixture was stirred at 80°C for 3 hours. The reaction mixture was filtered and washed with toluene (100 ml). The filtrate was washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a yellow solid was obtained. The obtained solid was washed with boiling ethanol (50 ml) and a yellow solid (5.2 g, 87%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.4 to 7.6 (3H, m), 7.6 to 7.9 (4H, m), 7.98 (1H, dd, J=8 Hz, 2 Hz), 8.2 to 8.4 (3H, m), 8.50 (1H, d, J=2 Hz)

(2) Synthesis of 2-phenyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-9,10-dihydroanthracene

[0096] Under an atmosphere of argon, 4-(2,2-diphenylvinyl)bromobenzene (5.0 g, 15 mmole, 2.6 eq) was dissolved into a mixed solvent composed of anhydrous toluene (25 ml) and anhydrous THF (25 ml) and the resultant solution was cooled at -40°C in a dry ice/methanol bath. To the cooled solution, a hexane solution of n-butyllithium (1.60 mole/liter, 10 ml, 16 mmole, 1.1 eq) was added and the resultant mixture was stirred at -20°C for 1 hour. To the obtained mixture, 2-phenylanthraquinone (1.6 g, 5.6 mmole) was added and the resultant mixture was stirred at -20°C for 1 hour and at the room temperature for 7 hours and left standing for one night. To the obtained reaction mixture, a saturated aqueous solution of ammonium chloride (50 ml) was added and the reaction was deactivated. The formed organic layer was separated by filtration, washed with a saturated aqueous solution of sodium chloride (30 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a yellow oil was obtained. The obtained oil was purified by the column chromatography (silica gel/hexane+50% dichloromethane, dichloromethane and hexane+1% methanol, successively) and a white amorphous solid (2.3 g, 52%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 2.75 (1H, s), 2.78 (1H, s), 6.68 (8H, s), 6.83 (2H, s), 7.1 to 7.7 (31H, m), 7.90 (1H, d, J=2 Hz)

(3) Synthesis of 3-phenyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-anthracene (Compound 34)

[0097] 2-Phenyl-9,10-bis(4-(2,2-diphenylvinyl)phenyl)-9,10-dihydroanthracene (2.3 g, 2.9 mmole) was dissolved into acetic acid (20 ml). To the resultant solution, potassium iodide (1.4 g, 8.4 mmole, 3 eq) and sodium phosphinate monohydrate (0.4 g, 3.8 mmole) were added and the obtained solution was stirred for 1 hours. The reaction mixture was filtered and washed with water, methanol and acetone and a light yellow solid (2.1 g, 95%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.14 (2H, s), 7.2 to 7.5 (39H, m), 7.87 (1H, s)

[0098] The solid obtained above (2.1 g) was purified by sublimation at 370°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (0.9 g) was obtained.

FDMS: calc. for C₆₀H₄₂=762, found m/z=762 (M⁺, 100)

λ_{max}, 409, 388, 370 nm (PhMe)

F_{max}, 453 nm (PhMe, λ_{ex}=409 nm)

E_g=2.85 eV

I_p=5.70 eV(14 Y/eV, 100 nW)

T_g=114°C

Synthesis Example 13 (Compound 41)

(1) Synthesis of 9-(4-bromophenyl)anthraquinone

[0099] Under an atmosphere of argon, 9-anthraceneboronic acid (11.8 g, 53 mmole), 4-iodobromobenzene (16.5 g, 58 mmole, 1.1 eq) and tetrakis(triphenylphosphine)palladium(0) (1.0 g, 0.87 mmole, 1.5% Pd) were dissolved in toluene (160 ml). To the obtained solution, a 2 M aqueous solution of sodium carbonate (17 g, 0.16 mole, 3 eq/80 ml) was added and the resultant solution was heated under refluxing for 10 hours. The formed organic layer was separated, washed with a 5% aqueous solution of sodium hydroxide (100 ml) and a saturated aqueous solution of sodium chloride (50 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a light yellow solid was obtained. The obtained solid was purified by recrystallization from toluene (30 ml) and white plate crystals (10 g, 57%)

were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.2 to 7.8 (10H, m), 8.0 to 8.1 (2H, m), 8.49 (1H, s)

(2) Synthesis of 9-bromo-10-(4-bromophenyl)anthracene

[0100] 9-(4-Bromophenyl)anthracene (6.3 g, 19 mmole) was dissolved in anhydrous dimethylformamide (DMF) (100 ml). To the resultant solution, a solution (15 ml) of NBS (3.7 g, 21 mmole, 1.1 eq) in anhydrous DMF was added and the obtained mixture was stirred at the room temperature for 7 hours and left standing for one night. After the reaction mixture was diluted with water (30 ml), the formed solid was separated by filtration and washed with methanol and a yellow solid (6.0 g, 77%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.2 to 7.8 (10H, m), 8.60 (2H, d, J=9 Hz)

(3) Synthesis of 9-(4-(2,2-diphenylvinyl)phenyl)-10-(4-(4-(2,2-diphenylvinyl)phenyl)phenyl)anthracene (Compound 41)

[0101] Under an atmosphere of argon, 9-bromo-10-(4-bromophenyl)-anthracene (3.0 g, 7.3 mmole), 4-(2,2-diphenylvinyl)phenylboronic acid (5.7 g, 19 mmole, 2.6 eq) and tetrakis(triphenylphosphine)palladium(0) (0.34 g, 0.29 mmole, 2% Pd) were suspended into toluene (60 ml). To the obtained suspension, a 2 M aqueous solution of sodium carbonate (6.0 g, 57 mmole, 3 eq/30 ml) was added and the resultant mixture was heated under refluxing for 10 hours. The reaction mixture was filtered and washed with toluene, water and methanol and a green solid (4.5 g) was obtained. The obtained solid was suspended in boiling toluene (50 ml), cooled while being left standing, filtered and washed with toluene and a light green solid (3.9 g, 70%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.0 to 7.5 (34H, m), 7.6 to 7.8 (8H, m)

[0102] The solid obtained above (3.9 g) was purified by sublimation at 380°C under 10⁻⁶ Torr for 2 hours and a light yellow solid (3.3 g) was obtained.

FDMS: calc. for C₆₀H₄₂=762, found m/z=762 (M⁺, 100), 381 (M²⁺, 7)

λ_{max}, 397, 378, 357, 323 nm (PhMe)

Fmax, 442 nm (PhMe, λ_{ex}=397 nm)

Eg=2.95 eV

Ip=5.62 eV(32 Y/eV, 100 nW)

Tg=120°C

Synthesis Example 14 (Compound 42)

(1) Synthesis of 9-(3-(2,2-diphenylvinyl)phenyl)-10-(4-(3-(2,2-diphenylvinyl)phenyl)phenyl)anthracene (Compound 42)

[0103] Under an atmosphere of argon, 9-bromo-10-(4-bromophenyl)-anthracene (3.0 g, 7.3 mmole), 4-(2,2-diphenylvinyl)phenylboronic acid (6.6 g, 22 mmole, 3 eq) and tetrakis(triphenylphosphine)palladium(0) (0.34 g, 0.29 mmole, 2% Pd) were suspended into toluene (60 ml). To the obtained suspension, a 2 M aqueous solution of sodium carbonate (7.0 g, 66 mmole, 3 eq/35 ml) was added and the resultant mixture was heated under refluxing for 10 hours. The reaction mixture was filtered and washed with toluene, water and methanol and a gray solid (1.4 g, the first crop, 25%) was obtained. From the filtrate, the organic layer was separated, washed with a saturated aqueous solution of sodium chloride and dried with magnesium sulfate. The solvent was removed by distillation and a deep brown oil was obtained. When the obtained oil was dissolved in dichloromethane, crystals were formed soon. The formed crystals were separated by filtration and washed with a mixed solvent composed of hexane and dichloromethane and a white solid (3.3 g, the second crop, 59%) was obtained. The obtained crude crystals (3.5 g) were suspended in boiling toluene (40 ml), cooled while being left standing, filtered and washed with toluene and a light yellow solid (2.4 g) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.1 to 7.6 (32H, m) all-H

[0104] The solid obtained above (2.4 g) was purified by sublimation at 380°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (1.9 g) was obtained.

[0105] FDMS: calc. for C₆₀H₄₂=762, found m/z=762 (M⁺, 100), 508 (imp, 2), 381 (M²⁺, 7)

λ_{max}, 397, 377, 357 nm (PhMe)

Fmax, 423, 436 nm (PhMe, λ_{ex}=397 nm)

Eg=3.00 eV

Ip=5.77 eV(17 Y/eV, 100 nW)

Tg=108°C

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Synthesis Example 15 (Compound 43)

(1) Synthesis of 9-(4-formylphenyl)anthracene

[0106] Under an atmosphere of argon, 9-bromoanthracene (3.9 g, 15 mmole), 4-formylphenylboronic acid (2.5 g, 17 mmole, 1.1 eq), potassium fluoride (3.2 g, 56 mmole, 3 eq) and tris(dibenzylideneacetone)-dipalladium(0) (0.07 g, 76 mmole, 1% Pd) were suspended in anhydrous THF (25 ml). To the obtained suspension, a toluene solution of tri-t-butylphosphine (66%, 0.06 ml, 0.2 mmole, 1.3 eq to Pd) was added and the resultant mixture was heated under refluxing for 10 hours. To the reaction mixture, water (50 ml) and toluene (150 ml) were added. The formed organic layer was separated, washed with a saturated aqueous solution of sodium chloride (50 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a light yellow solid was obtained. The obtained solid was purified by the column chromatography (silica gel/ hexane+50% dichloromethane) and a light yellow solid (3.3 g, 78%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.3 to 7.7 (8H, m), 8.0 to 8.1 (2H, m), 8.10 (2H, d, J=8 Hz.), 8.52 (1H, s), 10.18 (1H, s)

(2) Synthesis of 9-bromo-10-(4-formylphenyl)anthracene

[0107] 9-(4-Formylphenyl)anthracene (3.3 g, 12 mmole) was suspended in anhydrous DMF (40 ml). To the resultant suspension, a solution (8 ml) of NBS (2.3 g, 13 mmole, 1.1 eq) in anhydrous DMF was added and the obtained mixture was stirred at the room temperature for 10 hours and left standing for one night. After the reaction mixture was diluted with water (50 ml), the solid was separated by filtration and washed with methanol and a yellow solid (3.9 g, 90%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.3 to 7.7 (8H, m), 8.10 (2H, d, J=8 Hz), 8.62 (2H, dd, J=8 Hz, 2 Hz), 10.19 (1H, s)

(3) Synthesis of 9-bromo-10-(4-(2-phenylvinyl)phenyl)anthracene

[0108] Under an atmosphere of argon, 9-bromo-10-(4-formylphenyl)-anthracene (3.9 g, 11 mmole) and diethyl benzylphosphonate (3 g, 13 mmole, 1.2 eq) were suspended into DMSO (25 ml). To the resultant suspension, potassium t-butoxide (1.6 g, 14 mmole, 1.1 eq) was added and the obtained mixture was stirred at the room temperature for 10 hours and left standing for one night. The reaction mixture was diluted with water (50 ml) and subjected to extraction with toluene (300 ml). The organic layer was washed with water (50 ml) and a saturated aqueous solution of sodium chloride (50 ml) and dried with magnesium sulfate. Then, the solvent was removed by distillation and a yellow solid was obtained. The obtained solid was purified by recrystallization from toluene (40 ml) and yellow needle crystals (4.1 g, 86%) were obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.2 to 7.8 (17H, m), 8.60 (2H, d, J=8 Hz)

(4) Synthesis of 9-(4-(2,2-diphenylvinyl)phenyl)-10-(4-(2-phenylvinyl)-phenyl)anthracene (Compound 43)

[0109] Under an atmosphere of argon, 9-bromo-10-(4-(2-phenylvinyl)-phenyl)anthracene (3.1 g, 7.1 mmole), 4-(2,2-diphenylvinyl)phenylboronic acid (2.4 g, 8.0 mmole, 1.1 eq) and tetrakis(triphenylphosphine)-palladium(0) (0.16 g, 0.14 mmole, 2% Pd) were suspended into toluene (25 ml). To the obtained suspension, a 2 M aqueous solution of sodium carbonate (2.5 g, 24 mmole, 3 eq/12 ml) was added and the resultant mixture was heated under refluxing for 10 hours. The reaction mixture was filtered and washed with water and methanol and a yellow solid was obtained. The obtained solid was suspended in boiling toluene (50 ml), cooled while being left standing, filtered and washed with toluene and a light yellow solid (3.8 g, 88%) was obtained.

¹H-NMR (CDCl₃, TMS) δ: 7.14 (2H, s), 7.3 to 7.8 (32H, m)

[0110] The solid obtained above (3.8 g) was purified by sublimation at 340°C under 10⁻⁶ Torr for 1 hour and a light yellow solid (2.9 g) was obtained.

FDMS: calc. for C₄₈H₃₄=610, found m/z=610 (M⁺, 100), 305 (M²⁺, 5)

λ_{max}, 398, 379, 360, 313, 304 nm (PhMe)

Fmax, 445 nm (PhMe, λ_{ex}=397 nm)

Eg=2.94 eV

I_p=5.68 eV(12 Y/eV, 100 nW)

Example 1

[0111] A glass substrate (manufactured by GEOMATEC Company) of 25 mm×75 mm×1.1 mm thickness having an ITO transparent electrode was cleaned by application of ultrasonic wave in isopropyl alcohol for 5 minutes and then

5 by exposure to ozone generated by ultraviolet light for 30 minutes. The glass substrate having the transparent electrode lines which had been cleaned was attached to a substrate holder of a vacuum vapor deposition apparatus. On the surface of the cleaned substrate at the side having the transparent electrode, a film of N,N'-bis(N,N'-diphenyl-4-aminophenyl)-N,N'-diephenyl-4,4'-diamino-1,1'-biphenyl (referred to as TPD232, hereinafter) having a thickness of 60 nm was formed so that the formed film covered the transparent electrode. The formed film of TPD232 worked as the first hole injecting layer (the hole transporting layer). Then, on the formed film of TPD232, a film of 4,4'-bis[N-(1-naphthyl)-N-phenylamino]-biphenyl (referred to as NPD, hereinafter) having a thickness of 20 nm was formed. The formed film of NPD worked as the second hole injecting layer (the hole transporting layer). On the formed film of NPD, Compound (15) was vacuum vapor deposited so that a film having a thickness of 40 nm was formed. The film of compound (15) worked as the light emitting layer. On the film formed above, a film of tris(8-quinolinol)aluminum (referred to as Alq, hereinafter) having a thickness of 20 nm was formed. The film of Alq worked as the electron injecting layer. Thereafter, Li (the source of lithium: manufactured by SAES GETTERS Company) and Alq were binary vapor deposited and an Alq:Li film was formed as the electron injecting layer (the cathode). On the formed Alq:Li film, metallic aluminum was vapor deposited to form a metal cathode and an organic EL device was prepared. When a direct current voltage of 6 V was applied to the organic EL device prepared above, blue light was emitted at a luminance of 80 cd/m², a maximum luminance of 23,000 cd/m² and an efficiency of light emission of 2.0 cd/A.

10

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[0112] The spectrum of the light emitted from this organic EL device is shown in Figure 1. As shown in Figure 1, the light emitted from this organic EL device had the peak wavelength at 450 nm and exhibited excellent purity of color.

[0113] Compound 15 had a glass transition temperature of 118°C and exhibited excellent heat resistance. When the obtained organic EL device was kept at a high temperature (85°C, 500 hours), no change was found in the properties and the excellent heat resistance was confirmed.

[0114] When the device was driven under a constant current at an initial luminance of 80 cd/m², the half-life was as long as 13,000 hours.

25 Examples 2 to 14

[0115] Organic EL devices were prepared in accordance with the same procedures as those conducted in Example 1 except that the compounds shown in Table 1 were used in place of Compound (15). A direct current voltage of 6 V was applied to the prepared organic EL devices. The luminance of the emitted light and the efficiency of light emission were measured and the color of the emitted light was observed. The results are shown in Table 1.

Table 1

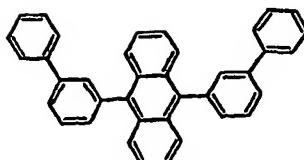
	Compound	Luminance of emitted light (cd/m ²)	Efficiency of light emission (cd/A)	Color of emitted light
35	Example 2 (1)	120	3.2	blue
	Example 3 (2)	113	2.7	blue
	Example 4 (3)	90	3.7	blue
	Example 5 (13)	130	2.2	blue
	Example 6 (14)	113	2.7	blue
	Example 7 (17)	90	4.2	blue
	Example 8 (20)	150	2.8	blue
	Example 9 (22)	180	4.7	blue
	Example 10 (25)	80	2.8	blue
	Example 11 (26)	75	2.6	blue
	Example 12 (33)	230	3.6	blue
	Example 13 (34)	280	4.3	blue
	Example 14 (41)	250	4.2	blue

50

Examples 15 to 25 and Comparative Examples 1 and 2

[0116] The glass transition temperatures of the novel compounds of the present invention shown in Table 2 and Compound C1 (Comparative Example 1):

55

5
10
15
20
?)
? 25
45
50
55
(C1)**(C2)**

and Compound C2 (Comparative Example 2):

were obtained by the measurement of DSC. The results are shown in Table 2.

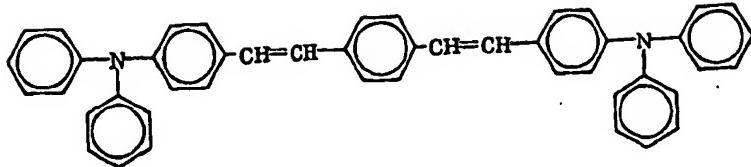
Table 2

	Compound	Glass transition temperature (°C)
Example 15	(14)	102
Example 16	(15)	118
Example 17	(17)	163
Example 18	(18)	106
Example 19	(22)	110
Example 20	(24)	113
Example 21	(25)	130
Example 22	(26)	135
Example 23	(33)	105
Example 24	(34)	110
Example 25	(41)	120
Comparative Example 1	(C1)	75
Comparative Example 2	(C2)	97

[0117] As shown in Table 2, the compounds of Comparative Examples had glass transition temperatures lower than 100°C and exhibited poor heat resistance. In contrast, the compounds of Examples had glass transition temperatures higher than 100°C and exhibited excellent heat resistance.

Example 26

[0118] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), Compound (15) and the following compound PAVB:



which worked as the fluorescent dopant were binary vapor deposited at a relative rate of vapor deposition of 40:1 and a film was formed. Under the application of a direct current voltage of 5.5 V to the prepared organic EL devices, the luminance of emitted light, the efficiency of light emission and the maximum luminance of emitted light were measured and the color of the emitted light was observed. The results are shown in Table 3.

Example 27

[0119] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), Compound (17) and PAVB shown above were binary vapor deposited at a relative rate of vapor deposition of 40:1 and a film was formed. Under the application of a direct current voltage of 5.5 V to the prepared organic EL devices, the luminance of emitted light, the efficiency of light emission and the maximum luminance of emitted light were measured and the color of the emitted light was observed. The results are shown in Table 3.

Example 28

[0120] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), Compound (18) and PAVB shown above were binary vapor deposited at a relative rate of vapor deposition of 40:1 and a film was formed. Under the application of a direct current voltage of 5.5 V to the prepared organic EL devices, the luminance of emitted light, the efficiency of light emission and the maximum luminance of emitted light were measured and the color of the emitted light was observed. The results are shown in Table 3.

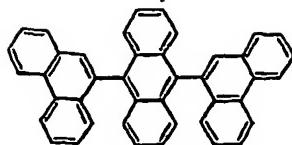
Table 3

	Compound	Luminance of emitted light	Efficiency of light emission	Maximum luminance of emitted light	Color of emitted light
		(cd/m ²)	(cd/A)	(cd/m ²)	
Example 26	(15) and PAVB	222	7.14	85,000	greenish blue
Example 27	(17) and PAVB	135	7.58	75,000	greenish blue
Example 28	(18) and PAVB	145	9.67	95,000	greenish blue

[0121] As shown in Table 3, the efficiency was improved by adding the fluorescent dopant to the novel compound of the present invention.

Comparative Example 3

[0122] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), the following Compound (C3):

5
(C3)

10 was vapor deposited and a film was formed. When a direct current voltage of 6.5 V was applied to the organic EL device prepared above, bluish green light was emitted at a luminance of 92 cd/m² and an efficiency of light emission of 1.22 cd/A. The efficiency was low and the device could not be used for a practical application.

15 Example 29

[0123] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that Compound (25) was used in place of Compound (15). When the device prepared above was driven under a constant current at an initial luminance of 500 cd/m², the half-life was as long as 840 hours which corresponded to about 6,000 hours at an initial luminance of 100 cd/m². When the organic EL device was kept at a high temperature (85°C, 500 hours), no change was found in the properties and the heat resistance was excellent. The light emission on the light emitting surface was uniform and showed no defects. The device exhibited an efficiency of light emission of 2.8 cd/A and blue light of a high purity having color coordinates of (0.16, 0.08) was emitted.

25 Example 30

[0124] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that Compound (33) was used in place of Compound (15). When the device prepared above was driven under a constant current at an initial luminance of 500 cd/m², the half-life was as long as 1,100 hours which corresponded to about 8,000 hours at an initial luminance of 100 cd/m². When the organic EL device was kept at a high temperature (85°C, 500 hours), no change was found in the properties and the heat resistance was excellent. The light emission on the light emitting surface was uniform and showed no defects. The device exhibited an excellent efficiency of light emission of 3.6 cd/A.

35 Example 31

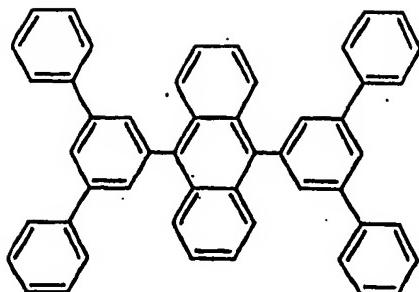
[0125] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that Compound (41) was used in place of Compound (15). When the device prepared above was driven under a constant current at an initial luminance of 500 cd/m², the half-life was as long as 1,200 hours which corresponded to about 9,500 hours at an initial luminance of 100 cd/m². When the organic EL device was kept at a high temperature (85°C, 500 hours), no change was found in the properties and the heat resistance was excellent. The light emission on the light emitting surface was uniform and showed no defects. Blue light of a high purity having color coordinates of (0.15, 0.13) was emitted and the device exhibited an excellent efficiency of light emission of 4.2 cd/A.

45 Comparative Example 4

[0126] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), the following Compound (C4):

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(C4)



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was vapor deposited and a film was formed. When the device prepared above was driven under a constant current at an initial luminance of 500 cd/m², the half-life was as extremely short as 25 hours and the device could not be used for practical applications. The efficiency of light emission was as low as 1.7 cd/A.

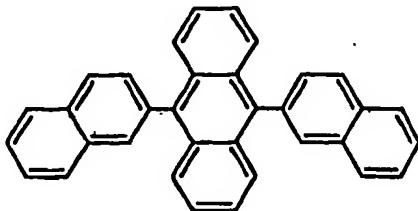
20 Comparative Example 5

[0127] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), the following Compound (C5):

25

30

(C5)



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was vapor deposited and a film was formed. When the device prepared above was driven under a constant current at an initial luminance of 500 cd/m², the half-life was as short as 420 hours and the device could not be used for practical applications. The efficiency of light emission was as low as 2.1 cd/A.

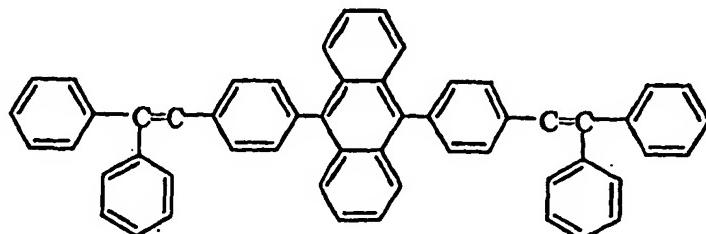
40 Comparative Example 6

[0128] An organic EL device was prepared in accordance with the same procedures as those conducted in Example 1 except that, in place of Compound (15), the following Compound (C6):

45

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(C6)



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was vapor deposited and a film was formed. When this device prepared above was kept at a high temperature (85°C, 500 hours), defects were formed at portions of the light emitting surface and the portions of the defects showed change

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in the color although the half-life was as long as 1,000 hours when the device was driven under a constant current at an initial luminance of 500 cd/m².

[0129] As described above, the devices prepared by using the compounds of the present invention could emit blue light at an efficiency of light emission of 2 cd/A or higher and were more excellent than the devices of Comparative Examples. The devices prepared by using the compounds of the present invention also had long lives, exhibited excellent heat resistance and could maintain uniform light emission after being kept at a high temperature.

INDUSTRIAL APPLICABILITY

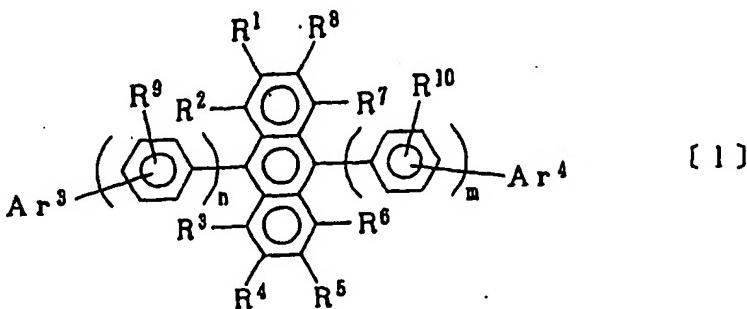
[0130] As described above in detail, the organic electroluminescence device of the present invention which utilizes any of the above novel compounds represented by general formulae [1], [1'] and [2] to [5] exhibits excellent efficiency of light emission and heat resistance, has a long life and emits bluish light having excellent purity of color.

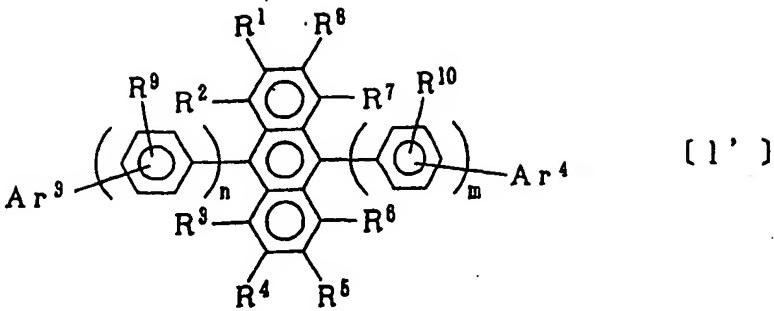
[0131] Therefore, the organic electroluminescence device of the present invention is useful as a light source such as a planar light emitting member of wall televisions and a back light of displays.

15

Claims

1. A novel compound represented by following general formula [1]:

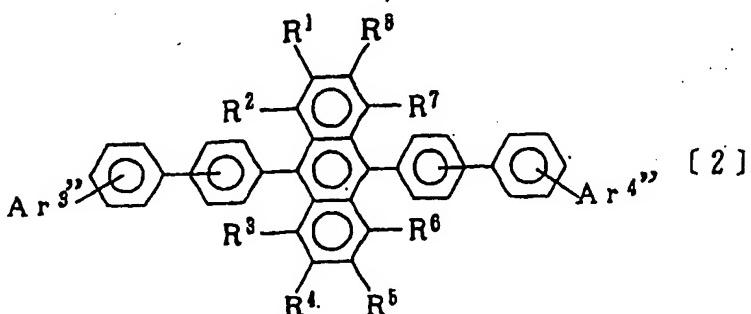




5 wherein R¹ to R¹⁰ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a
 10 substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an
 15 unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

20 Ar³ and Ar⁴ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 2 to 10 carbon atoms, the substituent in the above groups being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an
 25 unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms; and
 30 n represents a number of 1 to 3, m represents a number of 1 to 3 and a case in which n and m represent a same number is excluded.

35 3. A novel compound represented by following general formula [2]:



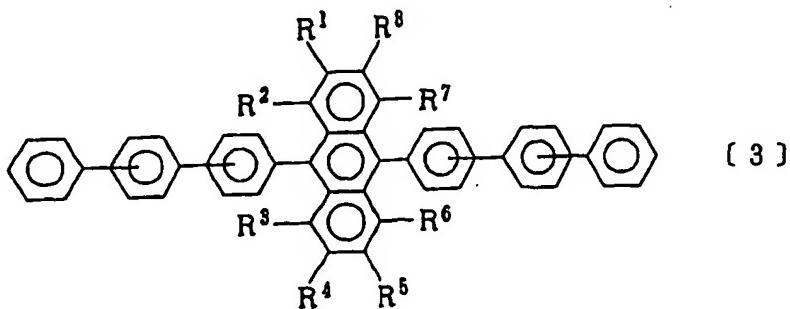
45 wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an
 50 unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

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atoms; and

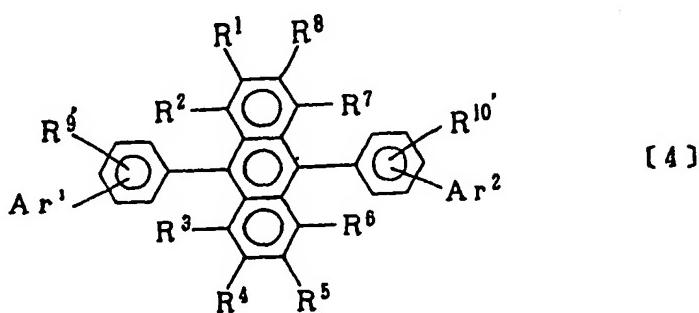
Ar^{3"} and Ar^{4"} each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, the substituent in the above group being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 6 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms, a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms or a substituted or unsubstituted alkenyl group having 4 to 40 carbon atoms.

4. A novel compound represented by following general formula [3]:



wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms.

- 30
- 35
5. A novel compound represented by following general formula [4]:



55

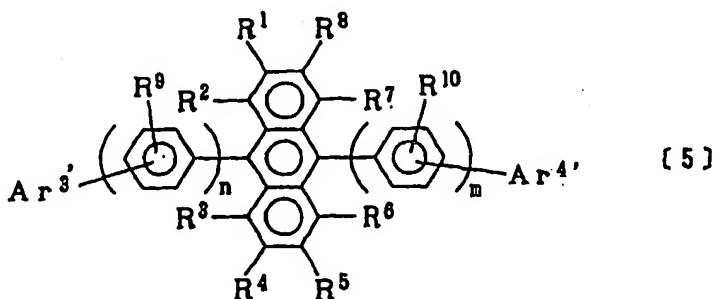
wherein R¹ to R⁸ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an

unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms;

5 R⁹ and R¹⁰ each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; and

10 Ar¹ and Ar¹ each independently represent a substituted or unsubstituted aryl group having 6 to 30 carbon atoms, the substituent in the above group being a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms.

- 15 6. A novel compound represented by following general formula [5]:



30 wherein R¹ to R¹⁰ each independently represent hydrogen atom, a halogen atom, cyano group, nitro group, a substituted or unsubstituted alkyl group having 1 to 20 carbon atoms, a substituted or unsubstituted alkoxy group having 1 to 20 carbon atoms, a substituted or unsubstituted aryloxy group having 6 to 30 carbon atoms, a substituted or unsubstituted alkylthio group having 1 to 20 carbon atoms, a substituted or unsubstituted arylthio group having 6 to 30 carbon atoms, a substituted or unsubstituted arylalkyl group having 7 to 30 carbon atoms, an unsubstituted monocyclic group having 5 to 30 carbon atoms, a substituted or unsubstituted condensed polycyclic group having 10 to 30 carbon atoms or a substituted or unsubstituted heterocyclic group having 5 to 30 carbon atoms, and at least one of R¹ to R⁸ represents the alkyl group, the alkoxy group, the aryloxy group, the alkylthio group, the arylthio group, the arylalkyl group, the monocyclic group, the condensed polycyclic group or the heterocyclic group;

35 Ar^{3'} and Ar^{4'} each independently represent a substituted or unsubstituted alkenyl group having 8 to 30 carbon atoms; and

40 n represents a number of 1 or 2 and m represents a number of 1 or 2.

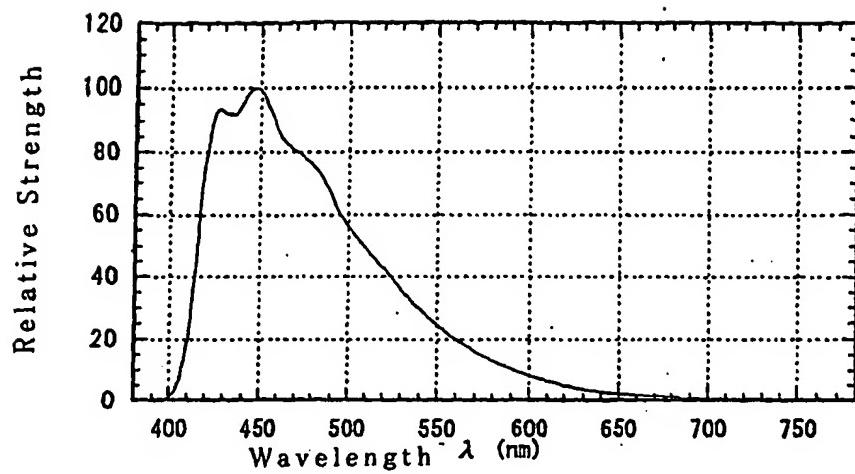
- 45 7. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein at least one of the layers of thin films of organic compounds comprises a novel compound described in Claim 1.
- 50 8. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein at least one of the layers of thin films of organic compounds comprises a novel compound described in Claim 2.
- 55 9. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein at least one of the layers of thin films of organic compounds comprises a novel compound described in Claim 3.

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- 5 23. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 5 and a fluorescent dopant.
- 10 24. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 6 and a fluorescent dopant.
- 15 25. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 1 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 20 26. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 2 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 25 27. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 3 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 30 28. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 4 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 35 29. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 5 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 40 30. An organic electroluminescence device comprising a plurality of layers of thin films of organic compounds which comprise a light emitting layer or a plurality of layers comprising a light emitting layer and are disposed between a pair of electrodes, wherein the light emitting layer comprises a novel compound described in Claim 6 and the organic electroluminescence device emits light having a peak wavelength of 460 nm or shorter.
- 45 31. An organic electroluminescence device according to Claim 19, wherein the fluorescent dopant is an amine compound.
- 50 32. An organic electroluminescence device according to Claim 20, wherein the fluorescent dopant is an amine compound.
- 55 33. An organic electroluminescence device according to Claim 21, wherein the fluorescent dopant is an amine compound.
- 60 34. An organic electroluminescence device according to Claim 22, wherein the fluorescent dopant is an amine compound.
- 65 35. An organic electroluminescence device according to Claim 23, wherein the fluorescent dopant is an amine compound.
- 70 36. An organic electroluminescence device according to Claim 24, wherein the fluorescent dopant is an amine compound.

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Fig. 1



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP01/09659

A. CLASSIFICATION OF SUBJECT MATTER		
Int.C1' C07C15/20, 15/56, C07D333/16, C09K11/06, H05B33/14		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED		
Minimum documentation searched (classification system followed by classification symbols) Int.C1' C07C15/20, 15/56, C07D333/16, C09K11/06, H05B33/14		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) CAPLUS (STN), REGISTRY (STN)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 1009041 A2 (Eastman Kodak Company), 14 June, 2000 (14.06.2000)	1, 7, 13, 19, 25, 31
A	& JP 2000-182775 A & US 2001-51285 A1 Claims; working example; chemical compounds No.5-17, 54	2-6, 8-12, 14-18, 20-24, 26-30, 32-36
X	JP 2000-143569 A (Sansei SDI Co., Ltd.), 23 May, 2000 (23.05.2000)	1, 7, 13, 19, 25, 31
A	& KR 2000-32066 A Claims; working example; formula 8	2-6, 8-12, 14-18, 20-24, 26-30, 32-36
X	JP 2000-53677 A (Idemitsu Kosan Co., Ltd.), 22 February, 2000 (22.02.2000) (Family: none)	1, 6-7, 12-13, 18-19, 24-25,
A	Claims; working example; page 5, lines 3, 5 to 6, 8; page 6, lines 2, 6; page 7, lines 3 to 4	30-31, 36 2-5, 8-11, 14-17, 20-23, 26-29, 32-35
X	JP 2000-7604 A (Idemitsu Kosan Co., Ltd.), 11 January, 2000 (11.01.2000) (Family: none)	1-2, 6-8, 12-14, 18-20,
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "B" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed		
Date of the actual completion of the international search 04 March, 2002 (04.03.02)	Date of mailing of the international search report 12 March, 2002 (12.03.02)	
Name and mailing address of the ISA/ Japanese Patent Office	Authorized officer	
Facsimile No.	Telephone No.	

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP01/09659

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	Claims; working example; chemical compounds Nos. 1-2, 6, 9, 11-12, 14, 18, 20, 23-24, 28-30, 32-34, 38	24-26, 30-32, 36 3-5, 9-11, 15-17, 21-23, 27-29, 33-35
X	JP 11-323323 A (Toppan Printing Co., Ltd.), 26 November, 1999 (26.11.1999) (Family: none) Claims; working example; pages 5 to 6	1, 6-7, 12-13, 18-19, 24-25, 30-31, 36
A	Claims; working example; chemical compound 42	2-5, 8-11, 14-17, 20-23, 26-29, 32-35
X	JP 8-12969 A (TDK Corporation), 16 January, 1996 (16.01.1996) (Family: none)	1, 7, 13, 19, 25, 31
A	Claims; working example; chemical compound 42	2-6, 8-12, 14-18, 20-24, 26-30, 32-36
X	EP 681019 A2 (TDK Corporation), 08 November, 1995 (08.11.1995) & JP 8-12600 A & US 5635308 A	1-3, 6-9, 12-15, 18-21, 24-27, 30-33, 36
A	Claims; working example; chemical compounds Nos. I, II, III, IV, V, VI, VII, VIII, IX, X	4-5, 10-11, 16-17, 22-23, 28-29, 34-35

(19)日本国特許庁 (JP)

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C 0 9 K 11/06	6 1 0	C 0 9 K 11/06	6 1 0
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(32)優先日	平成11年11月8日 (1999.11.8)
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(33)優先権主張国	日本 (JP)

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(74)代理人	100078732 弁理士 大谷 保

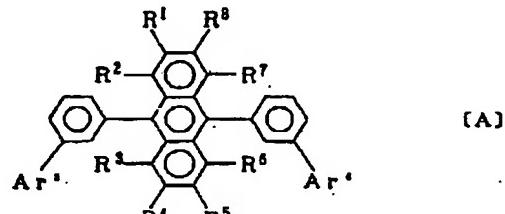
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(54)【発明の名称】 有機エレクトロルミネッセンス素子

(57)【要約】 (修正有)

【課題】 発光効率と、耐熱性に優れ、寿命が長く、色純度が良い青色系に発光する新規化合物及びそれを利用した有機エレクトロルミネッセンス(EL)素子を提供する。

【解決手段】 中心にジフェニルアントラセン構造、末端にアリール基置換された特定構造を有する一般式A、例えば式15の新規化合物。また一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなり、有機化合物薄膜の少なくとも一層が上記新規化合物を含有する有機EL素子。



(15)

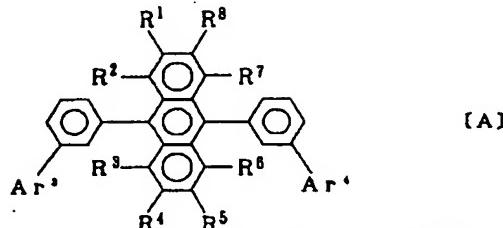


【特許請求の範囲】

【請求項1】 下記一般式〔A〕で示される新規化合物。

一般式〔A〕

〔化1〕

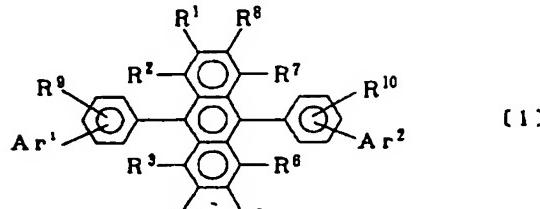


〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar³及びAr⁴は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、R⁹及びR¹⁰が水素原子かつAr¹及びAr²が未置換のフェニル基である場合と、R⁹及びR¹⁰がメタ位に置換するアリール基かつAr¹及びAr²がメタ位に置換するアリール基である場合を除く。〕

【請求項2】 下記一般式〔1〕で示される新規化合物。

一般式〔1〕

〔化2〕



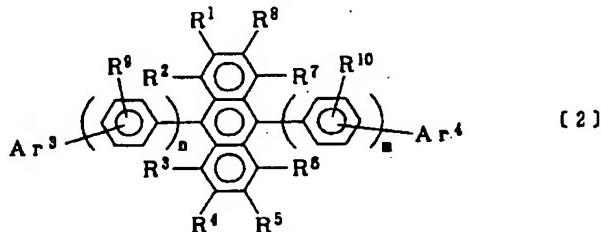
〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハ

ロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar¹及びAr²は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、R⁹及びR¹⁰が水素原子かつAr¹及びAr²が未置換のフェニル基である場合と、R⁹及びR¹⁰がメタ位に置換するアリール基かつAr¹及びAr²がメタ位に置換するアリール基である場合を除く。〕

【請求項3】 下記一般式〔2〕で示される新規化合物。

一般式〔2〕

〔化3〕



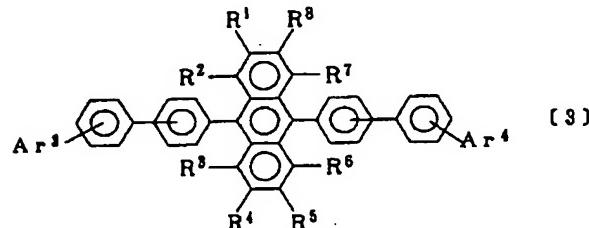
〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。ただし、Ar³及びAr⁴が未置換のフェニル基である場合を除く。〕

に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。nは1～3、mは1～3である。】

【請求項4】 下記一般式〔3〕で示される新規化合物。

一般式〔3〕

【化4】



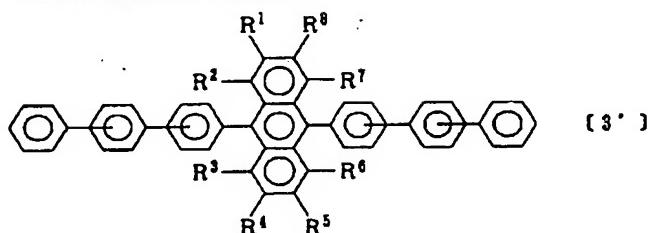
〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。nは1～3、mは1～3である。〕

換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar³及びAr⁴は、それ各自立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。〕

【請求項5】 下記一般式〔3'〕で示される新規化合物。

一般式〔3'〕

【化5】

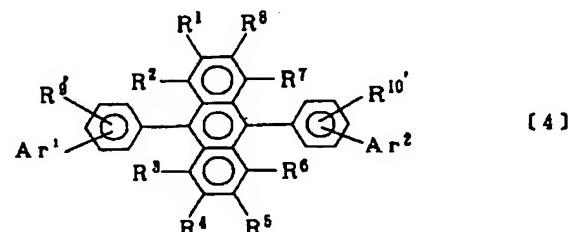


〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。〕

【請求項6】 下記一般式〔4〕で示される新規化合物。

一般式〔4〕

【化6】



〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。〕

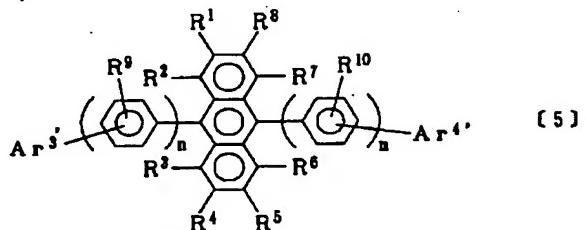
環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。R^{9'}及びR^{10'}は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。Ar¹及びAr²は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換も

しくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。】

【請求項7】 下記一般式〔5〕で示される新規化合物。

一般式〔5〕

【化7】



〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、R¹～R⁸の少なくとも一つは、アルキル基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アリールアルキル基、単環基、縮合多環基又は複素環基である。Ar^{3'}及びAr^{4'}は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。nは1～2、mは1～2である。〕

【請求項8】 一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、該有機化合物薄膜の少なくとも一層が請求項1～7のいずれかに記載の新規化合物を含有する層であることを特徴とする有機エレクトロルミネッセンス素子。

【請求項9】 一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、該発光層が請求項1～7のいずれかに記載の新規化合物を含有する層であることを特徴とする有機エレクトロルミネッセンス素子。

【請求項10】 一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレク

トロルミネッセンス素子において、該発光層が請求項1～7のいずれかに記載の新規化合物と蛍光性のドーパントとを含有する層であることを特徴とする有機エレクトロルミネッセンス素子。

【請求項11】 一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、該発光層が請求項1～7のいずれかに記載の新規化合物を含有する層であり、ピーク波長が460 nm以下の発光をすることを特徴とする有機エレクトロルミネッセンス素子。

【請求項12】 前記蛍光性のドーパントがアミン系化合物であることを特徴とする請求項10に記載の有機エレクトロルミネッセンス素子。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明は壁掛けテレビの平面発光体やディスプレイのバックライト等の光源として使用され、発光効率が高く、耐熱性に優れ、寿命が長く、色純度が良い青色系に発光する新規化合物及びそれを利用した有機エレクトロルミネッセンス素子に関するものである。

【0002】

【従来の技術】 有機物質を使用した有機エレクトロルミネッセンス(EL)素子は、固体発光型の安価な大面积フルカラー表示素子としての用途が有望視され、多くの開発が行われている。一般にEL素子は、発光層および該層をはさんだ一対の対向電極から構成されている。発光は、両電極間に電界が印加されると、陰極側から電子が注入され、陽極側から正孔が注入される。さらに、この電子が発光層において正孔と再結合し、励起状態を生成し、励起状態が基底状態に戻る際にエネルギーを光と

して放出する現象である。従来の有機EL素子は、無機発光ダイオードに比べて駆動電圧が高く、発光輝度や発光効率も低かった。また、特性劣化も著しく実用化には至っていなかった。最近の有機EL素子は徐々に改良されているものの、未だ充分な発光効率、耐熱性、寿命を有していなかった。例えば、特開平8-12600号公報にはフェニルアントラセンの2又は3量体の化合物を用いた有機EL素子が開示されているが、この化合物を利用した有機EL素子は、アントラセンを2又は3含み共役基で連結するため、エネルギーギャップが小さくなり、青色発光の色純度が劣っていた。また、この化合物は酸化しやすく不純物が存在し易く精製という点で問題があった。そこで、アントラセンの1, 9位にナフタレン置換した化合物やジフェニルアントラセンのフェニル基にm位アリール置換を行った化合物を用いた有機EL素子が試みられているが、発光効率が低く実用的ではなかった。また、特開平11-3782号公報には、ナフタレン置換したモノアントラセン誘導体を用いた有機EL素子が開示されている。しかしながら、発光効率が1cd/A程度と低く実用的ではなかった。さらに、米国特許第5972247号明細書には、フェニルアントラセン構造を有する化合物を用いた有機EL素子が開示されている。しかしながら、この化合物はm位にアリール置換を行っているため耐熱性は優れているものの、発光効率が2cd/A程度と低く実用的ではなかった。

【0003】

【発明が解決しようとする課題】本発明は、前記の課題を解決するためになされたもので、発光効率が高く、耐熱性に優れ、寿命が長く、色純度が良い青色系に発光する新規化合物及びそれを利用した有機エレクトロルミネッセンス素子を提供することを目的とするものである。

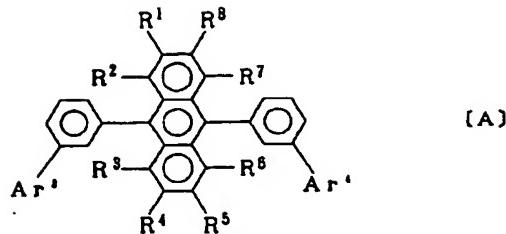
【0004】

【課題を解決するための手段】本発明者らは、前記の好ましい性質を有する新規化合物およびそれを使用した有機エレクトロルミネッセンス素子（以下、有機EL素子）を開発すべく鋭意研究を重ねた結果、中心にジフェニルアントラセン構造を持ち、末端にアリール基で置換された特定構造の新規化合物を利用することによりその目的を達成し得ることを見出した。本発明は、かかる知見に基づいて完成したものである。

【0005】すなわち、本発明の新規化合物は、下記一般式[A]、[1]～[5]及び[3']で示される化合物である。

【0006】一般式[A]

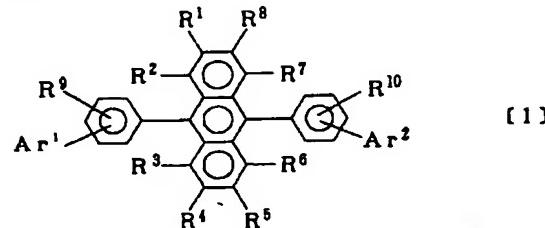
【化8】



〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar³及びAr⁴は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数6～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。ただし、Ar³及びAr⁴が未置換のフェニル基である場合を除く。〕

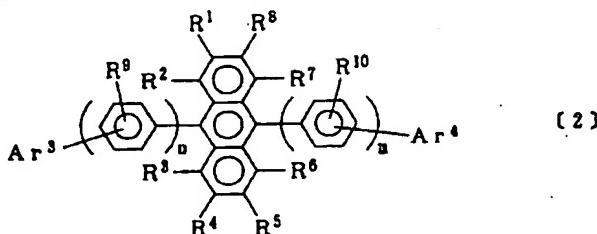
【0007】一般式[1]

【化9】



〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。〕

アリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar¹及びAr²は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ



〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar³及びAr⁴は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。nは1～3、mは1～3である。〕

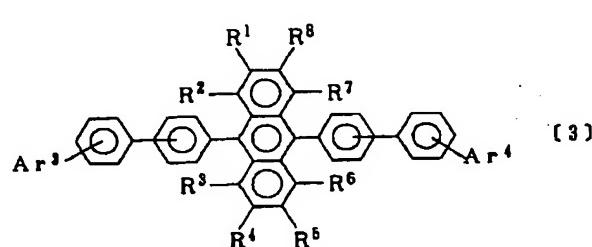
【0009】一般式〔3〕

【化11】

基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、R⁹及びR¹⁰が水素原子かつAr¹及びAr²が未置換のフェニル基である場合と、R⁹及びR¹⁰がメタ位に置換するアリール基かつAr¹及びAr²がメタ位に置換するアリール基である場合を除く。〕

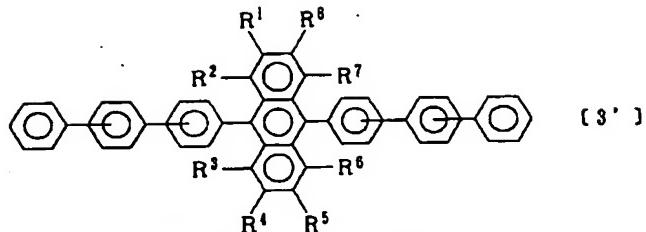
【0008】一般式〔2〕

【化10】



〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。Ar³及びAr⁴は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基、置換もしくは未置換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。〕

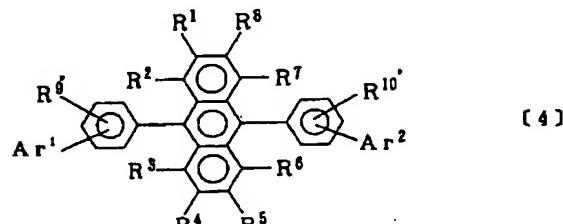
【0010】一般式〔3'〕



〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。〕

【0011】一般式〔4〕

【化13】

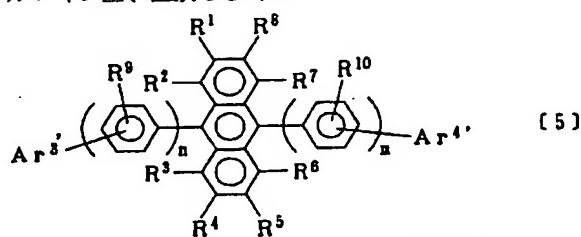


〔式中、R¹～R⁸は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは

未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。R^{9'}及びR^{10'}は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。Ar¹及びAr²は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。〕

【0012】一般式〔5〕

【化14】



〔式中、R¹～R¹⁰は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の単環基、置換もしくは未置換の炭素原子数10～30の縮

合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、R¹～R⁸の少なくとも一つは、アルキル基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アリールアルキル基、单環基、縮合多環基又は複素環基である。Ar^{3'}及びAr^{4'}は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。nは1～2、mは1～2である。〕

【0013】本発明の有機EL素子は、一対の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形

成してなる有機エレクトロルミネッセンス素子において、該有機化合物薄膜の少なくとも一層が、上記一般式[A]、[1]～[5]及び[3']で示されるいずれかの新規化合物を含有する。

【0014】前記発光層は、上記一般式[A]、[1]～[5]及び[3']で示されるいずれかの新規化合物を含有することが好ましい。前記発光層は、上記一般式[A]、[1]～[5]及び[3']で示されるいずれかの新規化合物と蛍光性のドーパントとを含有していてもよい。本発明の有機EL素子は、ピーク波長が460nm以下の発光であることが好ましい。前記蛍光性のドーパントは、アミン系化合物であることが好ましい。

【0015】

【発明の実施の形態】本発明の青色発光する有機EL素子に有用な新規化合物は、上記一般式[A]、[1]～[5]及び[3']のいずれかで示される。上記一般式[A]における $R^1 \sim R^8$ は、それぞれ独立に、水素原子、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。上記一般式[A]における A^{r3} 及び A^{r4} は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数6～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。上記一般式[A]における A^{r5} 及び A^{r6} は、それぞれ独立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数6～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。

換の炭素原子数5～30の複素環基又は置換もしくは未置換の炭素原子数4～40のアルケニル基である。ただし、 A^{r3} 及び A^{r4} が未置換のフェニル基である場合を除く。また、 A^{r3} 及び／又は A^{r4} がアリール基であった場合、炭素原子数10以上であるとガラス転移温度が高いため好ましい。

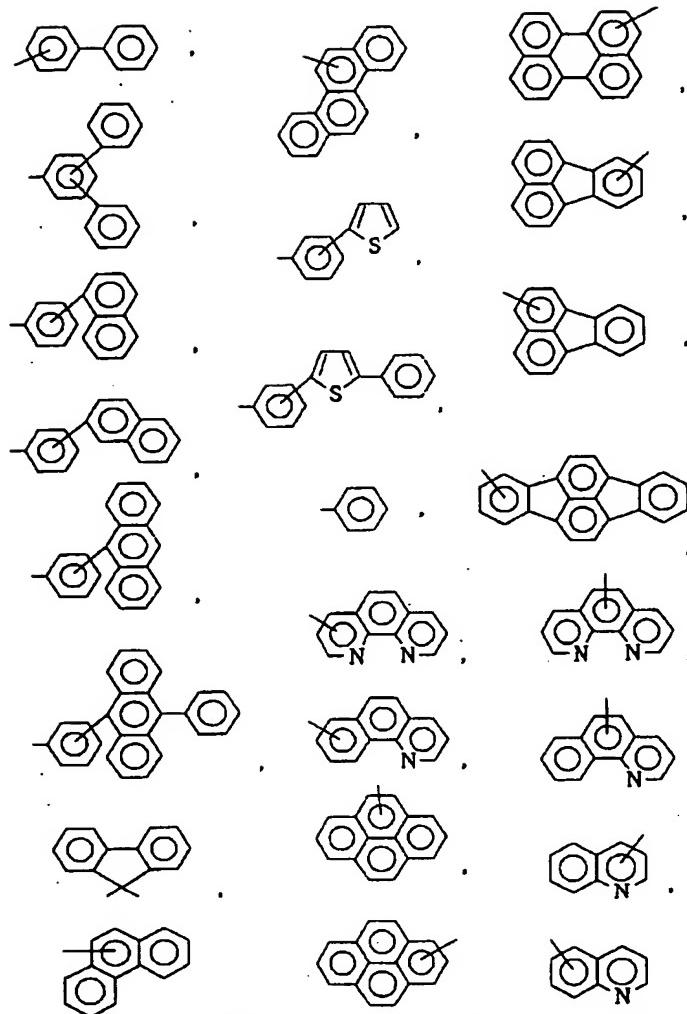
【0016】上記一般式[1]における $R^1 \sim R^{10}$ は、一般式[A]における $R^1 \sim R^8$ と同様である。上記一般式[1]における A^{r1} 及び A^{r2} は、それ各自立に、置換もしくは未置換の炭素原子数6～30のアリール基であり、置換基としては、置換もしくは未置換の炭素原子数1～20のアルキル基、置換もしくは未置換の炭素原子数1～20のアルコキシ基、置換もしくは未置換の炭素原子数6～30のアリールオキシ基、置換もしくは未置換の炭素原子数1～20のアルキルチオ基、置換もしくは未置換の炭素原子数6～30のアリールチオ基、置換もしくは未置換の炭素原子数7～30のアリールアルキル基、未置換の炭素原子数5～30の单環基、置換もしくは未置換の炭素原子数10～30の縮合多環基又は置換もしくは未置換の炭素原子数5～30の複素環基である。ただし、上記一般式[1]においては、 R^9 及び R^{10} が水素原子かつ A^{r1} 及び A^{r2} が未置換のフェニル基である場合と、 R^9 及び R^{10} がメタ位に置換するアリール基かつ A^{r1} 及び A^{r2} がメタ位に置換するアリール基である場合を除く。

【0017】上記一般式[2]における $R^1 \sim R^{10}$ は、一般式[A]における $R^1 \sim R^8$ と同様である。上記一般式[2]における A^{r3} 及び A^{r4} は、一般式[A]における A^{r3} 及び A^{r4} と同様である。上記一般式[2]におけるnは1～3、mは1～3である。

【0018】上記一般式[3]における $R^1 \sim R^8$ は、一般式[A]における $R^1 \sim R^8$ と同様である。上記一般式[3]における A^{r3} 及び A^{r4} は、一般式[A]における A^{r3} 及び A^{r4} と同様である。上記一般式[3']における $R^1 \sim R^8$ は、一般式[A]における $R^1 \sim R^8$ と同様である。

【0019】上記 $A^{r1} \sim A^{r4}$ として、例えば、

【化15】



及びこれらのアルキル置換体、アルコキシ置換体又はアリール置換体が挙げられる。

【0020】上記一般式〔4〕におけるR¹～R⁸は、一般式〔A〕におけるR¹～R⁸と同様であり、R⁹及びR¹⁰は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。上記一般式〔4〕におけるAr¹及びAr²は、一般式〔1〕におけるAr¹及びAr²と同様である。上記一般式〔4〕の化合物は、R⁹及びR¹⁰がアルケニル基であることから、有機EL素子に用いた場合、蛍光性が高くなり、発光効率が向上する。

【0021】上記一般式〔5〕におけるR¹～R¹⁰は、一般式〔A〕におけるR¹～R⁸と同様である。ただし、R¹～R⁸の少なくとも一つは、アルキル基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アリールアルキル基、単環基、縮合多環基又は複素環基である。上記一般式〔5〕におけるAr³及びAr⁴は、それぞれ独立に、置換もしくは未置換の炭素原子数8～30のアルケニル基である。上記一般式〔5〕の化合物は、R¹～R⁸の少なくとも一つが、ア

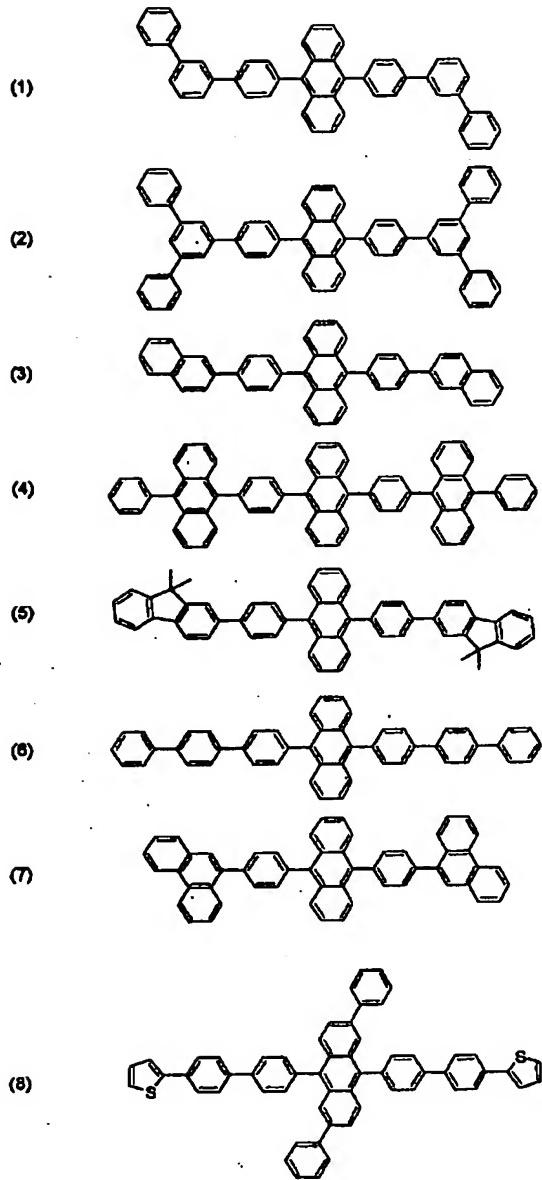
ルキル基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アリールアルキル基、単環基、縮合多環基又は複素環基であることから、有機EL素子に用いた場合、発光面の発光均一性が高くなり、欠陥が発生しにくくなる。

【0022】本発明の有機EL素子は、上記したように、一对の電極間に発光層または発光層を含む複数層の有機化合物薄膜を形成してなる有機エレクトロルミネッセンス素子において、該有機化合物薄膜の少なくとも一層が、上記一般式〔A〕、〔1〕～〔5〕及び〔3〕で示されるいずれかの新規化合物を含有する。前記有機化合物薄膜には、上記一般式〔A〕、〔1〕～〔5〕及び〔3〕で示されるいずれかの新規化合物が、1～100mol%含有されていることが好ましく、10～98mol%含有されているとさらに好ましい。本発明の有機EL素子は、青色系の発光をするものである。

【0023】以下に、本発明の一般式〔A〕、〔1〕～〔5〕及び〔3〕の化合物の代表例(1)～(40)を例示するが、本発明はこの代表例に限定されるものではない。

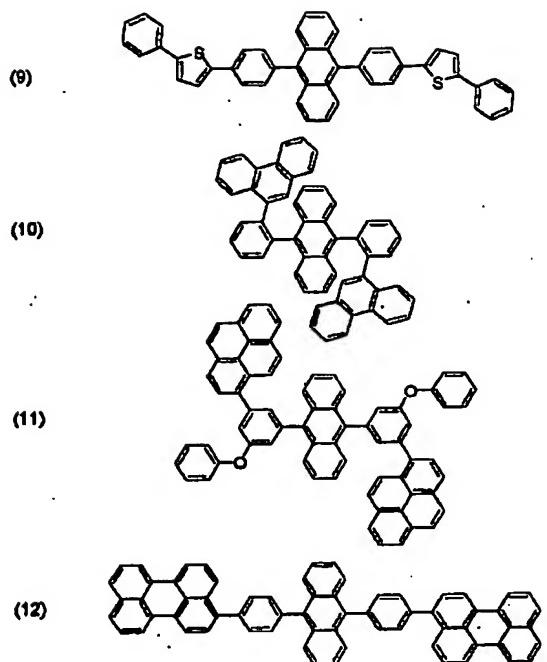
【0024】

【化16】



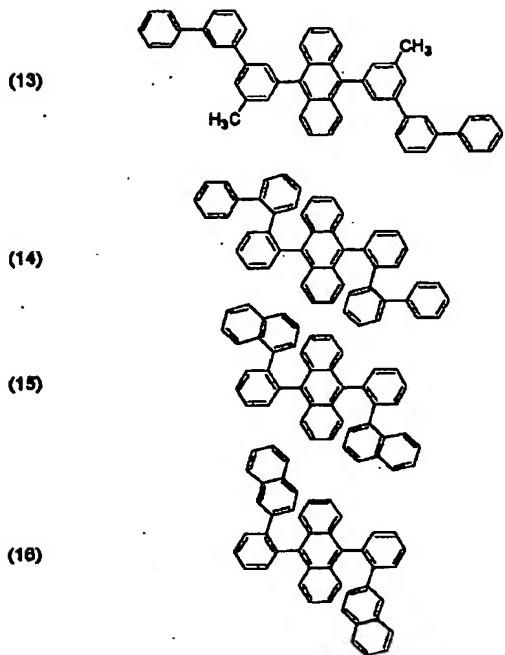
【0025】

【化17】



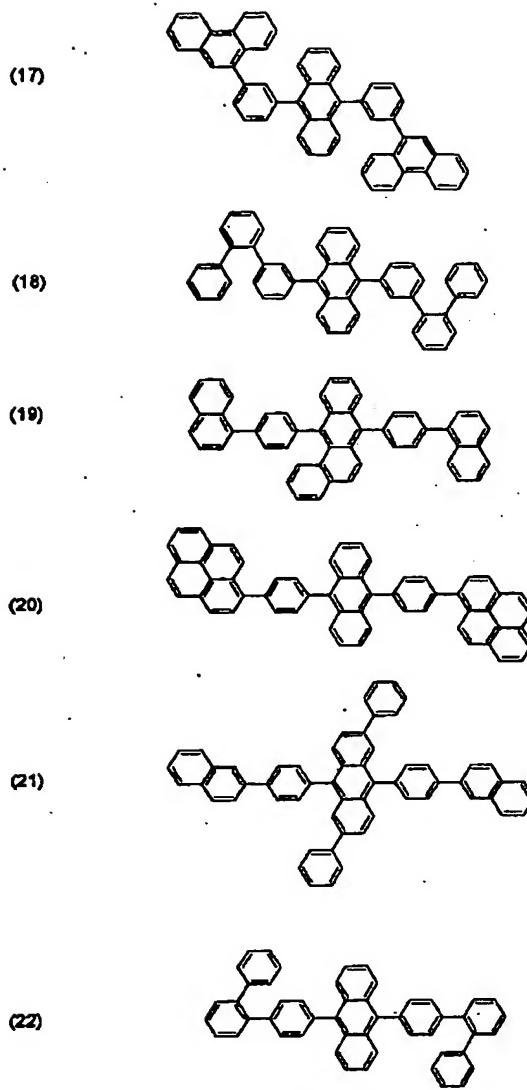
【0026】

【化18】



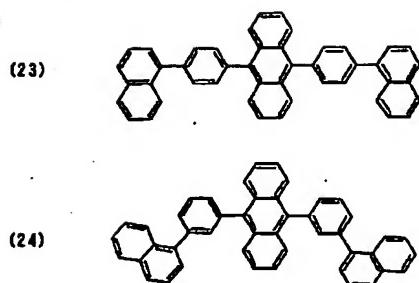
【0027】

【化19】



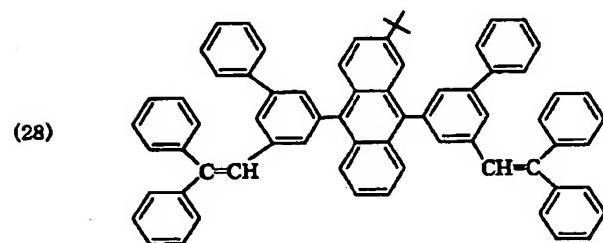
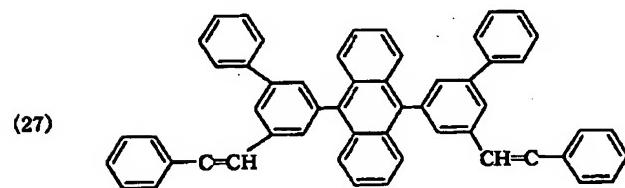
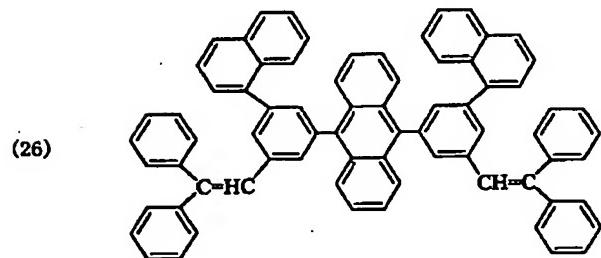
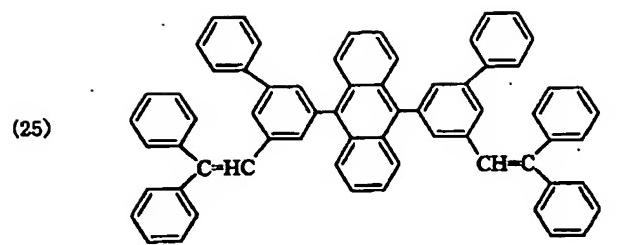
【0028】

【化20】



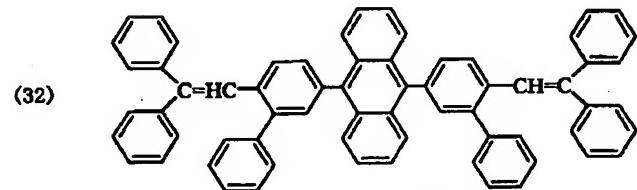
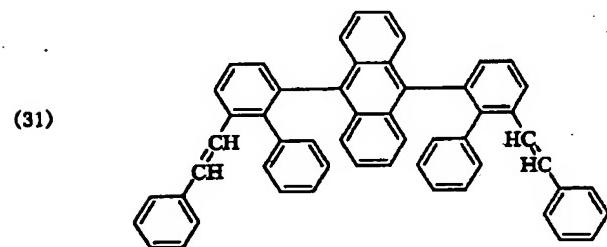
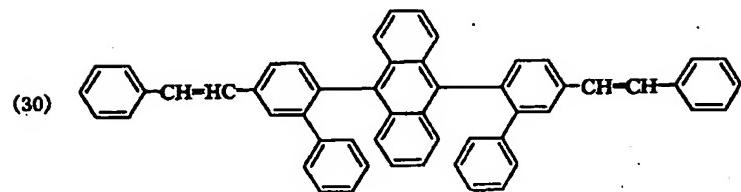
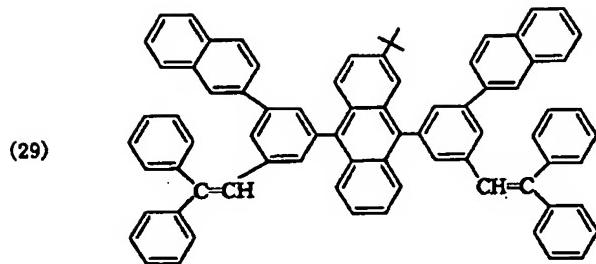
【0029】

【化21】



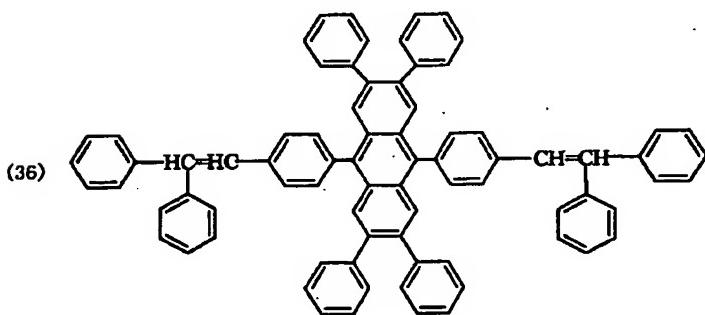
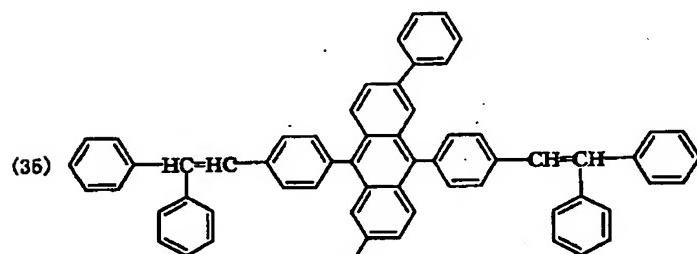
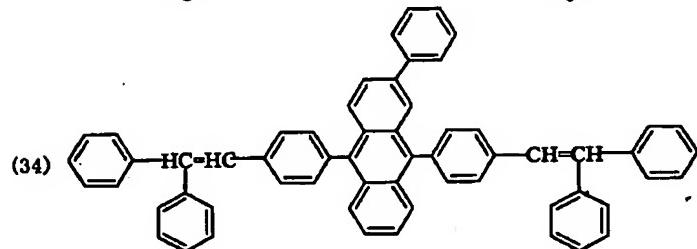
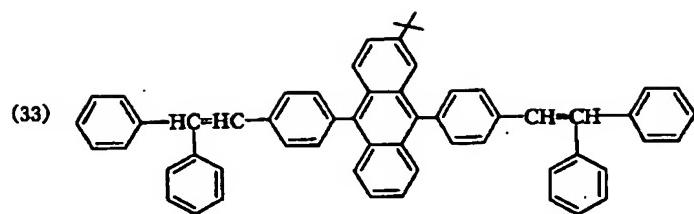
【0030】

【化22】



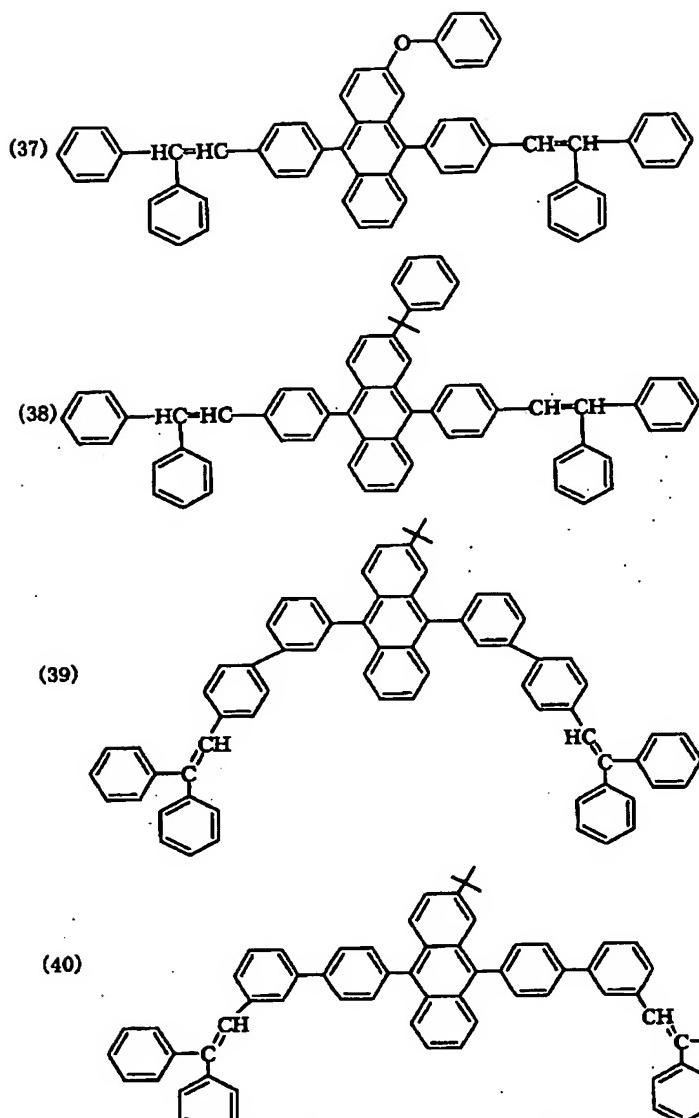
【0031】

【化23】



【0032】

【化24】



【0033】本発明の有機EL素子は、陽極と陰極間に一層もしくは多層の有機層を形成した素子である。一層型の場合、陽極と陰極との間に発光層を設けている。発光層は、発光材料を含有し、それに加えて陽極から注入した正孔、もしくは陰極から注入した電子を発光材料まで輸送させるために、正孔注入材料もしくは電子注入材料を含有しても良い。しかしながら、発光材料は、極めて高い蛍光量子効率、高い正孔輸送能力および電子輸送能力を併せ持ち、均一な薄膜を形成することが好ましい。多層型の有機EL素子は、(陽極/正孔注入層/発光層/陰極)、(陽極/発光層/電子注入層/陰極)、(陽極/正孔注入層/発光層/電子注入層/陰極)の多層構成で積層したものがある。

【0034】発光層には、必要に応じて、本発明の一般式[A]、[1]～[5]及び[3']のいずれかの化合物に加えてさらなる公知の発光材料、ドーピング材料、正孔注入材料や電子注入材料を使用することもでき

る。有機EL素子は、多層構造により、クエンチングによる輝度や寿命の低下を防ぐことができる。必要があれば、発光材料、他のドーピング材料、正孔注入材料や電子注入材料を組み合わせて使用することができる。また、他のドーピング材料により、発光輝度や発光効率の向上、赤色や白色の発光を得ることもできる。また、正孔注入層、発光層、電子注入層は、それぞれ二層以上の層構成により形成されても良い。その際には、正孔注入層の場合、電極から正孔を注入する層を正孔注入層、正孔注入層から正孔を受け取り発光層まで正孔を輸送する層を正孔輸送層と呼ぶ。同様に、電子注入層の場合、電極から電子を注入する層を電子注入層、電子注入層から電子を受け取り発光層まで電子を輸送する層を電子輸送層と呼ぶ。これらの各層は、材料のエネルギー準位、耐熱性、有機層もしくは金属電極との密着性等の各要因により選択されて使用される。

【0035】一般式[A]、[1]～[5]及び

〔3〕のいずれかの化合物と共に有機層に使用できる発光材料またはホスト材料としては、アントラセン、ナフタレン、フェナントレン、ビレン、テトラセン、コロネン、クリセン、フルオレセイン、ペリレン、フタロペリレン、ナフタロペリレン、ペリノン、フタロペリノン、ナフタロペリノン、ジフェニルブタジエン、テトラフェニルブタジエン、クマリン、オキサジアゾール、アルダジン、ビスベンゾキノリン、ビススチリル、ピラジン、シクロベンタジエン、キノリン金属錯体、アミノキノリン金属錯体、ベンゾキノリン金属錯体、イミン、ジフェニルエチレン、ビニルアントラセン、ジアミノカルバゾール、ピラン、チオピラン、ポリメチン、メロシアニン、イミダゾールキレート化オキソノイド化合物、キナクリドン、ルブレン、スチルベン系誘導体及び蛍光色素等が挙げられるが、これらに限定されるものではない。

【0036】正孔注入材料としては、正孔を輸送する能力を持ち、陽極からの正孔注入効果、発光層または発光材料に対して優れた正孔注入効果を有し、発光層で生成した励起子の電子注入層または電子注入材料への移動を防止し、かつ薄膜形成能力の優れた化合物が好ましい。具体的には、フタロシアニン誘導体、ナフタロシアニン誘導体、ポルフィリン誘導体、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、イミダゾロン、イミダゾールチオン、ピラゾリン、ピラゾロン、テトラヒドロイミダゾール、オキサゾール、オキサジアゾール、ヒドラゾン、アシルヒドラゾン、ポリアリールアルカン、スチルベン、ブタジエン、ベンジジン型トリフェニルアミン、スチリルアミン型トリフェニルアミン、ジアミン型トリフェニルアミン等と、それらの誘導体、およびポリビニルカルバゾール、ポリシラン、導電性高分子等の高分子材料が挙げられるが、これらに限定されるものではない。

【0037】本発明の有機EL素子において使用できる正孔注入材料の中で、さらに効果的な正孔注入材料は、芳香族三級アミン誘導体もしくはフタロシアニン誘導体である。芳香族三級アミン誘導体の具体例は、トリフェニルアミン、トリトリルアミン、トリルジフェニルアミン、N, N'-ジフェニル-N, N'-(3-メチルフェニル)-1, 1'-ビフェニル-4, 4'-ジアミン、N, N, N', N'-(4-メチルフェニル)-1, 1'-フェニル-4, 4'-ジアミン、N, N, N', N'-(4-メチルフェニル)-1, 1'-ビフェニル-4, 4'-ジアミン、N, N'-ジフェニル-N, N'-ジナフチル-1, 1'-ビフェニル-4, 4'-ジアミン、N, N'-メチルフェニル)-N, N'-(4-n-ブチルフェニル)-フェナントレン-9, 10-ジアミン、N, N-ビス(4-ジ-4-トリルアミノフェニル)-4-フェニル-シクロヘキサン等、もしくはこれらの芳香族三級アミン骨格を有したオ

リゴマーもしくはポリマーであるが、これらに限定されるものではない。フタロシアニン(Pc)誘導体の具体例は、H₂Pc、CuPc、CoPc、NiPc、ZnPc、PdPc、FePc、MnPc、ClAlPc、ClGaPc、ClInPc、ClSnPc、Cl₂SiPc、(HO)AlPc、(HO)GaPc、VOPc、TiOPc、MoOPc、GaPc-O-GaPc等のフタロシアニン誘導体およびナフタロシアニン誘導体であるが、これらに限定されるものではない。

【0038】電子注入材料としては、電子を輸送する能力を持ち、陰極からの電子注入効果、発光層または発光材料に対して優れた電子注入効果を有し、発光層で生成した励起子の正孔注入層への移動を防止し、かつ薄膜形成能力の優れた化合物が好ましい。具体的には、フルオレノン、アントラキノジメタン、ジフェノキノン、チオピランジオキシド、オキサゾール、オキサジアゾール、トリアゾール、イミダゾール、ペリレンテトラカルボン酸、フレオレニリデンメタン、アントラキノジメタン、アントロン等とそれらの誘導体が挙げられるが、これらに限定されるものではない。また、正孔注入材料に電子受容物質を、電子注入材料に電子供与性物質を添加することにより電荷注入性を向上させることもできる。

【0039】本発明の有機EL素子において、さらに効果的な電子注入材料は、金属錯体化合物もしくは含窒素五員環誘導体である。金属錯体化合物の具体例は、8-ヒドロキシキノリナートリチウム、ビス(8-ヒドロキシキノリナート)亜鉛、ビス(8-ヒドロキシキノリナート)銅、ビス(8-ヒドロキシキノリナート)マンガン、トリス(8-ヒドロキシキノリナート)アルミニウム、トリス(2-メチル-8-ヒドロキシキノリナート)アルミニウム、トリス(8-ヒドロキシキノリナート)ガリウム、ビス(10-ヒドロキシベンゾ[h]キノリナート)ベリリウム、ビス(10-ヒドロキシベンゾ[h]キノリナート)亜鉛、ビス(2-メチル-8-キノリナート)クロロガリウム、ビス(2-メチル-8-キノリナート)(o-クレゾラート)ガリウム、ビス(2-メチル-8-キノリナート)(1-ナフトラート)アルミニウム、ビス(2-メチル-8-キノリナート)(2-ナフトラート)ガリウム等が挙げられるが、これらに限定されるものではない。

【0040】また、含窒素五員誘導体は、オキサゾール、チアゾール、オキサジアゾール、チアジアゾールもしくはトリアゾール誘導体が好ましい。具体的には、2, 5-ビス(1-フェニル)-1, 3, 4-オキサゾール、ジメチルPOPPOP、2, 5-ビス(1-フェニル)-1, 3, 4-チアゾール、2, 5-ビス(1-フェニル)-1, 3, 4-オキサジアゾール、2-(4'-tert-ブチルフェニル)-5-(4"-ビフェニル)-1, 3, 4-オキサジアゾール、2, 5-ビス(1-ナフチル)-1, 3, 4-オキサジアゾール、1, 4-

ービス [2-(5-フェニルオキサジアゾリル)]ベンゼン、1,4-ビス[2-(5-フェニルオキサジアゾリル)-4-tert-ブチルベンゼン]、2-(4'-tert-ブチルフェニル)-5-(4"-ビフェニル)-1,3,4-チアジアゾール、2,5-ビス(1-ナフチル)-1,3,4-チアジアゾール、1,4-ビス[2-(5-フェニルチアジアゾリル)]ベンゼン、2-(4'-tert-ブチルフェニル)-5-(4"-ビフェニル)-1,3,4-トリアゾール、2,5-ビス(1-ナフチル)-1,3,4-トリアゾール、1,4-ビス[2-(5-フェニルトリニアゾリル)]ベンゼン等が挙げられるが、これらに限定されるものではない。

【0041】本発明の有機EL素子においては、有機層中に、一般式[A]、[1]～[5]及び[3']の化合物の他に、発光材料、ドーピング材料、正孔注入材料および電子注入材料の少なくとも1種が同一層に含有されてもよい。また、本発明により得られた有機EL素子の、温度、湿度、雰囲気等に対する安定性の向上のために、素子の表面に保護層を設けたり、シリコンオイル、樹脂等により素子全体を保護することも可能である。

【0042】有機EL素子の陽極に使用される導電性材料としては、4eVより大きな仕事関数を持つものが適しており、炭素、アルミニウム、バナジウム、鉄、コバルト、ニッケル、タンクステン、銀、金、白金、パラジウム等およびそれらの合金、ITO基板、NEESA基板に使用される酸化スズ、酸化インジウム等の酸化金属、さらにはポリチオフェンやポリピロール等の有機導電性樹脂が用いられる。陰極に使用される導電性物質としては、4eVより小さな仕事関数を持つものが適しており、マグネシウム、カルシウム、錫、鉛、チタニウム、イットリウム、リチウム、ルテニウム、マンガン、アルミニウム等およびそれらの合金が用いられるが、これらに限定されるものではない。合金としては、マグネシウム/銀、マグネシウム/インジウム、リチウム/アルミニウム等が代表例として挙げられるが、これらに限定されるものではない。合金の比率は、蒸着源の温度、雰囲気、真空度等により制御され、適切な比率を選択される。陽極および陰極は、必要があれば二層以上の層構成により形成されていても良い。

【0043】有機EL素子では、効率良く発光させるために、少なくとも一方の面は素子の発光波長領域において充分透明にすることが望ましい。また、基板も透明であることが望ましい。透明電極は、上記の導電性材料を使用して、蒸着やスパッタリング等の方法で所定の透光性が確保するように設定する。発光面の電極は、光透過率を10%以上にすることが望ましい。基板は、機械的、熱的強度を有し、透明性を有するものであれば限定されるものではないが、ガラス基板および透明性樹脂フィルムがある。透明性樹脂フィルムとしては、ポリエチ

レン、エチレン-酢酸ビニル共重合体、エチレン-ビニルアルコール共重合体、ポリプロピレン、ポリスチレン、ポリメチルメタアクリレート、ポリ塩化ビニル、ポリビニルアルコール、ポリビニルブチラール、ナイロン、ポリエーテルエーテルケトン、ポリサルホン、ポリエーテルサルフォン、テトラフルオロエチレン-バーフルオロアルキルビニルエーテル共重合体、ポリビニルフルオライド、テトラフルオロエチレン-エチレン共重合体、テトラフルオロエチレン-ヘキサフルオロプロピレン共重合体、ポリクロロトリフルオロエチレン、ポリビニリデンフルオライド、ポリエステル、ポリカーボネット、ポリウレタン、ポリイミド、ポリエーテルイミド、ポリイミド、ポリプロピレン等が挙げられる。

【0044】本発明に係わる有機EL素子の各層の形成は、真空蒸着、スパッタリング、プラズマ、イオンプレーティング等の乾式成膜法やスピンドルコーティング、ディップティング、フローコーティング等の湿式成膜法のいずれの方法を適用することができる。膜厚は特に限定されるものではないが、適切な膜厚に設定する必要がある。膜厚が厚すぎると、一定の光出力を得るために大きな印加電圧が必要になり効率が悪くなる。膜厚が薄すぎるとピンホール等が発生して、電界を印加しても充分な発光輝度が得られない。通常の膜厚は5nmから10μmの範囲が適しているが、10nmから0.2μmの範囲がさらに好ましい。

【0045】湿式成膜法の場合、各層を形成する材料を、エタノール、クロロホルム、テトラヒドロフラン、ジオキサン等の適切な溶媒に溶解または分散させて薄膜を形成するが、その溶媒はいずれであっても良い。また、いずれの有機薄膜層においても、成膜性向上、膜のピンホール防止等のため適切な樹脂や添加剤を使用しても良い。使用の可能な樹脂としては、ポリスチレン、ポリカーボネット、ポリアリレート、ポリエステル、ポリアミド、ポリウレタン、ポリスルファン、ポリメチルメタクリレート、ポリメチルアクリレート、セルロース等の絶縁性樹脂およびそれらの共重合体、ポリ-N-ビニルカルバゾール、ポリシラン等の光導電性樹脂、ポリチオフェン、ポリピロール等の導電性樹脂を挙げられる。また、添加剤としては、酸化防止剤、紫外線吸収剤、可塑剤等を挙げられる。

【0046】以上のように、有機EL素子の有機層に本発明の新規化合物を用いることにより、発光効率が高く、耐熱性に優れ、寿命が長く、色純度が良い青色系に発光する有機EL素子を得ることができる。本発明の有機EL素子は、壁掛けテレビのフラットパネルディスプレイ等の平面発光体、複写機、プリンター、液晶ディスプレイのバックライト又は計器類等の光源、表示板、標識灯等に利用できる。

【0047】

【実施例】以下、本発明を合成例及び実施例に基づいて

さらに詳細に説明する。

合成例1 (化合物14)

(1) 2, 2'-ジブロモビフェニルの合成

アルゴン雰囲気下、1, 2-ジブロモベンゼン(25g, 0.11mol)を無水THF(240ml)に溶かしドライアイス/メタノール浴で-67°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 35ml, 53mmol, 0.5eq)を5分かけて徐々に滴下した。-67°Cで1時間攪拌後、室温で3時間攪拌した。反応混合物に飽和塩化アンモニウム水溶液(100ml)を加えて、有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して褐色液体(ca. 19g)を得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン)で精製して白色針状晶(9.5g, 57%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.2-7.4 (6H, m), 7.6-7.7 (2H, m)

【0048】(2) 2-フェニル-2'-ブロモビフェニルの合成

アルゴン雰囲気下、2, 2'-ジブロモビフェニル(9.5g, 30mmol)、フェニルボロン酸(3.7g, 30mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(1.0g, 0.87mmol, 3% Pd)をトルエン(75ml)に溶かし、炭酸ナトリウム水溶液(9.7g, 92mmol, 3eq/46ml)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して暗黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色針状晶(5.4g, 83%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.6-6.7 (1H, m), 7.0-7.6 (12H, m)

【0049】(3) 9, 10-ビス(2-(2-フェニルフェニル)フェニル)-9, 10-ジヒドロキシー-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、2-フェニル-2'-ブロモビフェニル(6.5g, 21mmol, 2.5eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-30°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 15ml, 23mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.7g, 8.2mmol)を加えて-20°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(2.9g, 53%)を得た。

¹H-NMR(CDCl₃, TMS) δ 0.47 (2H, s), 5.7-5.8 (2H, m), 6.3-7.4 (30H, m), 8.3-8.4 (2H, m)

【0050】(4) 9, 10-ビス(2-(2-フェニルフェニル)フェニル)アントラセン(化合物14)の合成

9, 10-ビス(2-(2-フェニルフェニル)フェニル)-9, 10-ジヒドロキシー-9, 10-ジヒドロアントラセン(2.9g, 4.3mmol)を酢酸(45ml)に懸濁し、57% よう化水素酸(6ml, 45mmol, 10eq)を加えて100°Cで6時間攪拌した。反応混合物に50% 次亜りん酸水溶液(30ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(2.4g, 88%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (34H, m). all-H

【0051】合成例2 (化合物15)

(1) 1-(2-ブロモフェニル)ナフタレンの合成
アルゴン雰囲気下、2-ブロモヨードベンゼン(7.0g, 25mmol)、ナフタレンボロン酸(4.0g, 23mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.5g, 0.43mmol, 1.7% Pd)をトルエン(50ml)に溶かし、炭酸ナトリウム水溶液(7.3g, 69mmol, 3eq/35ml)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色針状晶(5.4g, 83%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.3-7.8 (9H, m), 7.90 (2H, d, J=8Hz, 2Hz)

【0052】(2) 9, 10-ビス(2-(1-ナフチル)フェニル)-9, 10-ジヒドロキシー-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1-(2-ブロモフェニル)ナフタレン(5.4g, 19mmol, 2.8eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 14ml, 21mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.4g, 6.7mmol)を加えて-20°Cで1時間、室温で3時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(3.5g, 85%)を得た。

¹H-NMR(CDCl₃, TMS) δ -0.20 (2H, s), 5.76 (2H, dd, J=13Hz, 7Hz), 6.2-7.7 (26H, m), 8.43 (2H, d, J=8Hz)

【0053】(3) 9, 10-ビス(2-(1-ナフチル)フェニル)アントラセン(化合物15)の合成

9, 10-ビス(2-(1-ナフチル)フェニル)-9, 10-ジヒドロキシー-9, 10-ジヒドロアントラセン(3.5g, 5.7mmol)を酢酸(80ml)に懸濁し、57% よう化水素酸(15ml, 0.11mol, 20eq)を加えて100°Cで7時間攪拌した。反応混合物に50% 次亜りん酸水溶液(30ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(3.2g, 96%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (30H, m). all-H

【0054】合成例3（化合物17）

（1）9-フェナントレンボロン酸の合成

アルゴン雰囲気下、9-ブロモフェナントレン(15g, 58mmol)を無水エーテル(150ml)に溶かし、ドライアイス/メタノール浴で-35°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 43ml, 65mmol)を滴下し、-20°Cで1時間攪拌した。反応混合物を-67°Cに冷却し、トリイソプロポキシボラン(37ml, 0.16mol, 2.8eq)の無水エーテル溶液(30ml)を加えて、-65°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に10% 塩酸水溶液(150ml)を加えて室温で1時間攪拌後、有機層を分取、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して得られた固体をヘキサンで洗浄して白色固体(10g, 78%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.6-7.9 (5H, m), 8.17 (1H, s), 8.5-8.8 (3H, m)

【0055】（2）9-(3-ブロモフェニル)フェナントレンの合成

アルゴン雰囲気下、3-ブロモヨードベンゼン(7.0g, 25mmol)、フェナントレンボロン酸(5.0g, 23mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.5g, 0.43mmol, 1.7% Pd)をトルエン(100ml)に溶かし、炭酸ナトリウム水溶液(7.3g, 69mmol, 3eq/35ml)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色針状晶(6.6g, 80%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.3-8.7 (11H, m), 8.76 (2H, d, J=7Hz)

【0056】（3）9,10-ビス(3-(9-フェナントリル)フェニル)-9,10-ジヒドロキシ-9,10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1-(3-ブロモフェニル)フェナントレン(6.5g, 20mmol, 2.8eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-25°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 15ml, 23mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.5g, 7.2mmol)を加えて-20°Cで1時間、室温で3時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(5.9g, quant)を得た。

¹H-NMR(CDCl₃, TMS) δ -0.16 (2H, s), 6.06 (2H, s), 6.4-7.0 (12H, m), 7.1-7.7 (12H, m), 8.20 (2H, dd, J=8Hz, 2Hz), 8.4-8.6 (6H, m)

【0057】（4）9,10-ビス(3-(9-フェナントリル)フェニル)アントラセン(化合物17)の合

成

9,10-ビス(3-(9-フェナントリル)フェニル)-9,10-ジヒドロキシ-9,10-ジヒドロアントラセン(5.2g, 7.3mmol)を酢酸(120ml)に懸濁し、57% よう化水素酸(10ml, 77mmol, 10eq)を加えて100°Cで6時間攪拌した。反応混合物に50% 次亜りん酸水溶液(40ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(5.0g, quant)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.0-8.5 (34H, m). all-H

【0058】合成例4（化合物20）

（1）1-(4-ブロモフェニル)ビレンの合成

アルゴン雰囲気下、4-ブロモヨードベンゼン(7.0g, 25mmol)、1-ビレンボロン酸(5.7g, 23mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.5g, 0.43mmol, 1.7% Pd)をトルエン(50ml)に溶かし、炭酸ナトリウム水溶液(7.3g, 69mmol, 3eq/35ml)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色針状晶(6.6g, 80%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.3-7.8 (11H, m), 7.90 (2H, d, J=8Hz)

【0059】（2）9,10-ビス(4-(1-ビレニル)フェニル)-9,10-ジヒドロキシ-9,10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1-(2-ブロモフェニル)ビレン(6.6g, 18mmol, 2.8eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 14ml, 21mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.4g, 6.7mmol)を加えて-20°Cで1時間、室温で3時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(4.5g, 88%)を得た。

¹H-NMR(CDCl₃, TMS) δ -0.20 (2H, s), 5.76 (2H, dd, J=13Hz, 7Hz), 6.2-7.7 (30H, m), 8.43 (2H, d, J=8Hz)

【0060】（3）9,10-ビス(4-(1-ビレニル)フェニル)アントラセン(化合物20)の合成

9,10-ビス(4-(1-ビレニル)フェニル)-9,10-ジヒドロキシ-9,10-ジヒドロアントラセン(4.5g, 5.9mmol)を酢酸(80ml)に懸濁し、57% よう化水素酸(15ml, 0.11mol, 20eq)を加えて100°Cで7時間攪拌した。反応混合物に50% 次亜りん酸水溶液(30ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(3.9g, 90%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (30H, m). a11-H

【0061】合成例5 (化合物22)

(1) 2-ビフェニルボロン酸の合成

アルゴン雰囲気下、2-ブロモビフェニル(20g, 86mmol)を無水エーテル(200ml)に溶かし、ドライアイス/メタノール浴で-35°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 63ml, 95mmol)を滴下し、-20°Cで1時間攪拌した。反応混合物を-67°Cに冷却し、トライソプロポキシボラン(50ml, 0.22mol, 2.5eq)の無水エーテル溶液(50ml)を加えて、-65°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に10% 塩酸水溶液(200ml)を加えて室温で1時間攪拌後、有機層を分取、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して得られた固体をヘキサンで洗浄して白色固体(11g, 62%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.74 (1H, d, J=7Hz), 7.1-7.4 (8H, m)

【0062】(2) 2-(4-ブロモフェニル)ビフェニルの合成

アルゴン雰囲気下、2-ブロモヨードベンゼン(7.9g, 25mmol)、ビフェニルボロン酸(5.0g, 25mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.5g, 0.43mmol, 1.7% Pd)をトルエン(60ml)に溶かし、炭酸ナトリウム水溶液(8.0g, 75mmol, 3eq/40ml)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、無水硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+5%ジクロロメタン)で精製して白色針状晶(6.8g, 88%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.3-8.7 (11H, m), 8.76 (2H, d, J=7Hz)

【0063】(3) 9, 10-ビス(4-(2-フェニルフェニル)フェニル)-9, 10-ジヒドロキシー9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、2-(4-ブロモフェニル)ビフェニル(6.8g, 22mmol, 2.5eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-30°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 15ml, 23mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.7g, 8.2mmol)を加えて-20°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(4.9g, 89%)を得た。

¹H-NMR(CDCl₃, TMS) δ 5.7-5.8 (2H, m), 6.3-7.4 (30H, m), 8.3-8.4 (2H, m)

【0064】(4) 9, 10-ビス(4-(2-フェニルフェニル)フェニル)アントラセン(化合物22)の

合成

9, 10-ビス(4-(2-フェニルフェニル)フェニル)-9, 10-ジヒドロアントラセン(4.9g, 7.3mmol)を酢酸(70ml)に懸濁し、57% よう化水素酸(10ml, 77mmol, 10eq)を加えて100 °Cで6時間攪拌した。反応混合物に50% 次亜りん酸水溶液(50ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(4.6g, 88%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (34H, m). a11-H

【0065】合成例6 (化合物1の合成)

(1) 9, 10-ビス(4-(3-フェニルフェニル)フェニル)-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、3-(4-ブロモフェニル)ビフェニル(6.8g, 22mmol, 2.5eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-30°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 15ml, 23mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.7g, 8.2mmol)を加えて-20°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(5.0g, 91%)を得た。

¹H-NMR(CDCl₃, TMS) δ 5.7-5.8 (2H, m), 6.3-7.4 (30H, m), 8.3-8.4 (2H, m)

【0066】(2) 9, 10-ビス(4-(3-フェニルフェニル)フェニル)アントラセン(化合物1)の合成
9, 10-ビス(4-(3-フェニルフェニル)フェニル)-9, 10-ジヒドロアントラセン(4.9g, 7.3mmol)を酢酸(70ml)に懸濁し、57% よう化水素酸(10ml, 77mmol, 10eq)を加えて100 °Cで6時間攪拌した。反応混合物に50% 次亜りん酸水溶液(50ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(4.1g, 79%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (34H, m). a11-H

【0067】合成例7 (化合物2の合成)

(1) 9, 10-ビス(4-(3, 5-ジフェニルフェニル)フェニル)-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1, 3-ジフェニル-5-(4-ブロモフェニル)ベンゼン(8.5g, 22mmol, 2.5eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-30°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 15ml, 23mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.7g, 8.2mmol)を加えて-20°Cで1時間、室温で2時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体を得た。

して白色固体(16g, 90%)を得た。

¹H-NMR(CDCl₃, TMS) δ 5.7-5.8 (2H, m), 6.3-7.4 (30H, m), 8.3-8.4 (2H, m)

【0068】(2) 9, 10-ビス(4-(3, 5-ジフェニルフェニル)フェニル)アントラセン(化合物2)の合成

9, 10-ビス(4-(3, 5-ジフェニルフェニル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセン(6.0g, 7.3mmol)を酢酸(70ml)に懸濁し、5% よう化水素酸(10ml, 77mmol, 10eq)を加えて100°Cで6時間攪拌した。反応混合物に50% 次亜りん酸水溶液(50ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(5.3g, 93%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (34H, m). a11-H

【0069】合成例8(化合物3の合成)

(1) 9, 10-ビス(4-(2-ナフチル)フェニル)-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、2-(4-ブロモフェニル)ナフタレン(5.4g, 19mmol, 2.8eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.50mol/l, 14ml, 21mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(1.4g, 6.7mmol)を加えて-20°Cで1時間、室温で3時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(20ml)を加えて生じた固体をろ別し、水、メタノール、アセトンで洗浄して白色固体(3.7g, 91%)を得た。

¹H-NMR(CDCl₃, TMS) δ -0.20 (2H, s), 5.76 (2H, dd, J=13Hz, 7Hz), 6.2-7.7 (26H, m), 8.43 (2H, d, J=8Hz)

【0070】(2) 9, 10-ビス(4-(2-ナフチル)フェニル)アントラセン(化合物3)の合成

9, 10-ビス(4-(2-ナフチル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセン(3.5g, 5.7mmol)を酢酸(80ml)に懸濁し、5% よう化水素酸(15ml, 0.11mol, 20eq)を加えて100°Cで7時間攪拌した。反応混合物に50% 次亜りん酸水溶液(30ml)を加え、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(3.3g, 98%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.7-7.5 (30H, m). a11-H

【0071】合成例9(化合物25)

(1) 1-(2, 2-ジフェニルビニル)-3, 5-ジブロモベンゼンの合成

アルゴン雰囲気下、3, 5-ジブロモベンズアルデヒド(12.1g, 46mmol)、ジフェニルメチルホスホン酸ジエチル(15g, 49mmol, 1.1eq)をDMSO(60ml)に溶かし、カリウムt-ブトキシド(6.2g, 55mmol, 1.2eq)を少量ずつ加えて、室温で9時間攪拌し、一晩放置した。反応混合

物に水(60ml)を加え、酢酸エチル(250ml)で抽出、有機層を水(100ml)、飽和食塩水(50ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して濃褐色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン)で精製して白色固体(14.0g, 74%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.80 (1H, s), 7.03 (2H, d, J=2Hz), 7.3-7.4 (11H, m)

【0072】(2) 1-(2, 2-ジフェニルビニル)-3-フェニル-5-ブロモベンゼンの合成

アルゴン雰囲気下、1-(2, 2-ジフェニルビニル)-3, 5-ジブロモベンゼン(7.0g, 17mmol)、フェニルボロン酸(2.1g, 17mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.4g, 0.35mmol, 2%Pd)をトルエン(40ml)に溶かし、2M炭酸ナトリウム水溶液(25ml, 51mmol, 3eq)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(30ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色針状晶(3.9g, 56%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.94 (1H, s), 7.1-7.5 (18H, m)

【0073】(3) 9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-フェニル)-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1-(2, 2-ジフェニルビニル)-3-フェニル-5-ブロモベンゼン(3.9g, 9.5mmol, 2.7eq)を無水トルエン(20ml)と無水THF(20ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.60mol/l, 6ml, 9.6mmol, 1.0eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(0.7g, 3.4mmol)を加え、-20°Cで1時間、室温で7時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(50ml)を加えて失活させ、有機層をろ取し、飽和食塩水(30ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン+50%ジクロロメタン、次にジクロロメタン、最後にジクロロメタン+3%メタノール)で精製して淡黄色アモルファス固体(2.0g, 67%)を得た。

¹H-NMR(CDCl₃, TMS) δ 2.56 (2H, s), 6.5-6.6 (4H, m), 6.8-7.4 (34H, m), 7.41 (4H, dd, J=6Hz, 3Hz), 7.71 (4H, dd, J=6Hz, 3Hz)

【0074】(4) 9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-フェニル)アントラセン(化合物25)の合成

9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-フェニル)-9, 10-ジヒドロキシー

9, 10-ジヒドロアントラセン(2.0g, 2.3mmol)を酢酸(25ml)に溶かし、よう化カリウム(1.5g, 90mmol, 4eq)を加えて3時間攪拌した。反応混合物に50% ホスフィン酸水溶液を加えて失活させ、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(1.4g, 73%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.2-7.4 (4H, m), 7.65 (4H, dd, J=7Hz, 3Hz)この固体(1.4g)を380 °C/10⁻⁶Torrで1時間昇華精製することにより淡黄色固体(0.8g)を得た。

FDMS, calcd for C₆₆H₄₆=838, found m/z=838(M⁺, 4)

λ_{max} 398, 358, 306nm(PhMe)

F_{max}, 416, 435nm(PhMe, λ_{ex}=395nm)

Eg=3.00eV

I_p=5.87eV(51Y/eV, 100nW)

T_g=130°C

【0075】合成例10(化合物26)

(1) 1-(2, 2-ジフェニルビニル)-3-(1-ナフチル)-5-ブロモベンゼンの合成

アルゴン雰囲気下、1-(2, 2-ジフェニルビニル)-3, 5-ジブロモベンゼン(8.3g, 20mmol)、1-ナフタレンボロン酸(3.4g, 20mmol)、テトラキス(トリフェニルホスフィン)パラジウム(0)(0.46g, 0.4mmol, 2%Pd)をトルエン(50ml)に溶かし、2M炭酸ナトリウム水溶液(30ml, 60mmol, 3eq)を加えて10時間還流し、一晩放置した。有機層を分取し、飽和食塩水(50ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン、次にヘキサン+3%ジクロロメタン、最後にヘキサン+10%ジクロロメタン)で精製して白色ガラス状固体(5.5g, 60%)を得た。

¹H-NMR(CDCl₃, TMS) δ 6.96 (1H, s), 7.1-7.6 (18H, m), 7.8-7.9 (2H, m)

【0076】(2) 9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-(1-ナフチル))-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、1-(2, 2-ジフェニルビニル)-3-(1-ナフチル)-5-ブロモベンゼン(5.5g, 12mmol, 2.7eq)を無水トルエン(30ml)と無水THF(30ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-30°Cに冷却した。これにn-ブチルリチウム/ヘキサン溶液(1.60mol/1, 8ml, 13mmol, 1.0eq)を加えて、-20°Cで1時間攪拌した。これにアントラキノン(0.9g, 4.4mmol)を加え、-20°Cで1時間、室温で7時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(50ml)を加えて失活させ、有機層をろ取し、飽和食塩水(30ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン+50%ジクロロメタン、次にジクロロメタン、最後にジクロロメタン+1%メタノール)で精製して白色アモルファス固体(3.3g, 75%)を得た。

クロロメタン)で精製して白色アモルファス固体(2.7g, 63%)を得た。

¹H-NMR(CDCl₃, TMS) δ 2.56 (2H, s), 6.5-6.8 (6H, m), 6.9-7.5 (36H, m), 7.6-7.8 (8H, m)

【0077】(3) 9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-(1-ナフチル))アントラセン(化合物26)の合成

9, 10-ビス(3-(2, 2-ジフェニルビニル)フェニル-5-(1-ナフチル))-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセン(2.7g, 2.8mmol)を酢酸(30ml)に溶かし、よう化カリウム(1.8g, 11mmol, 4eq)を加えて3時間攪拌した。反応混合物に50% ホスフィン酸水溶液(40ml)を加えて失活させ、固体をろ別、水、メタノール、アセトンで洗浄して白色固体(2.0g, 78%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.2-7.5 (40H, m), 7.7-7.9 (10H, m)

この固体(2.0g)を400 °C/10⁻⁶Torrで1時間昇華精製することにより淡黄色固体(1.2g)を得た。

FDMS, calcd for C₇₄H₅₀=938, found m/z=938(M⁺, 100), 469(M²⁺, 6)

λ_{max} 398, 377, 358nm(PhMe)

F_{max}, 418, 436nm(PhMe, λ_{ex}=395nm)

Eg=3.00eV

I_p=5.86eV(34Y/eV, 100nW)

T_g=132°C

【0078】合成例11(化合物33)

(1) 2-t-ブチル-9, 10-ビス(4-(2, 2-ジフェニルビニル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、4-(2, 2-ジフェニルビニル)ブロモベンゼン(5.0g, 15mmol, 2.6eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにブチルリチウム/ヘキサン溶液(1.60mol/1, 10ml, 16mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これに2-(t-ブチル)アントラキノン(1.5g, 5.7mmol)を加え、-20°Cで1時間、室温で7時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(50ml)を加えて失活させ、有機層をろ取し、飽和食塩水(30ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン+50%ジクロロメタン、次にジクロロメタン、最後にジクロロメタン+1%メタノール)で精製して白色アモルファス固体(3.3g, 75%)を得た。

¹H-NMR(CDCl₃, TMS) δ 1.29 (9H, s), 2.65 (1H, s), 2.71 (1H, s), 6.68 (9H, s), 6.84 (2H, s), 7.1-7.4 (23H, m), 7.5-7.7 (4H, m)

【0079】(2) 3-t-ブチル-9, 10-ビス(4-(2, 2-ジフェニルビニル)フェニル)アントラセンの合成

ラセン(化合物33)の合成

2-チアブチル-9, 10-ビス(4-(2, 2-ジフェニルビニル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセン(3.3g, 4.3mmol)を酢酸(30ml)に溶かし、よう化カリウム(1.9g, 11mmol, 2.7eq)ホスフィン酸ナトリウム1水和物(0.6g, 5.7mmol)を加えて2時間攪拌した。反応混合物をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(2.8g, 88%)を得た。

¹H-NMR(CDCl₃, TMS) δ 1.28(9H, s), 7.14(2H, s), 7.2-7.5(30H, m), 7.6-7.7(5H, m)

この固体(2.8g)を360 °C/10⁻⁶Torrで1時間昇華精製することにより淡黄色固体(2.2g)を得た。

FDMS, calcd for C₅₈H₄₆=742, found m/z=742(M⁺, 100), 371(M²⁺, 4)

λ_{max} 397, 379, 360, 310nm(PhMe)

F_{max}, 450nm(PhMe, λ_{ex}=397nm)

E_g=2.92eV

I_p=5.71eV(39Y/eV, 100nW)

T_g=105°C

【0080】合成例12(化合物34)

(1) 2-フェニルアントラキノンの合成

アルゴン雰囲気下、2-クロロアントラキノン(5.0g, 21mmol)、フェニルボロン酸(2.8g, 23mmol, 1.1eq)、トリス(ジベジリデンアセトン)ジバラジウム(0)(0.2g, 0.22mmol, 2%Pd)、フッ化カリウム(4.4g, 76mmol, 3.3eq)を無水ジオキサン(30ml)に懸濁させ、トリチアブチルホスフィン/トルエン溶液(66%, 0.13ml, 0.42mmol, 1eq)を加えて、80°Cで3時間攪拌した。反応混合物をろ別し、トルエン(100ml)で洗浄した。ろ液を飽和食塩水(30ml)で洗浄、硫酸マグネシウムで乾燥、溶媒留去して黄色固体を得た。これを沸騰エタノールで(50ml)で洗浄して黄色固体(5.2g, 87%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.4-7.6(3H, m), 7.6-7.9(4H, m), 7.98(1H, dd, J=8Hz, 2Hz), 8.2-8.4(3H, m), 8.50(1H, d, J=2Hz)

【0081】(2) 2-フェニル-9, 10-ビス(4-(2, 2-ジフェニルビニル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセンの合成

アルゴン雰囲気下、4-(2, 2-ジフェニルビニル)プロモベンゼン(5.0g, 15mmol, 2.6eq)を無水トルエン(25ml)と無水THF(25ml)の混合溶媒に溶かし、ドライアイス/メタノール浴で-40°Cに冷却した。これにブチルリチウム/ヘキサン溶液(1.60mol/l, 10ml, 16mmol, 1.1eq)を加えて、-20°Cで1時間攪拌した。これに2-フェニルアントラキノン(1.6g, 5.6mmol)を加え、-20°Cで1時間、室温で7時間攪拌し、一晩放置した。反応混合物に飽和塩化アンモニウム水溶液(50ml)を加えて失活させ、有機層をろ取し、飽和食塩水(30ml)で洗浄、硫酸

マグネシウムで乾燥、溶媒留去して黄色オイルを得た。これをカラムクロマトグラフィ(シリカゲル/ヘキサン+50%ジクロロメタン、次にジクロロメタン、最後にヘキサン+1%メタノール)で精製して白色アモルファス固体(2.3g, 52%)を得た。

¹H-NMR(CDCl₃, TMS) δ 2.75(1H, s), 2.78(1H, s), 6.68(8H, s), 6.83(2H, s), 7.1-7.7(31H, m), 7.90(1H, d, J=2Hz)

【0082】(3) 3-フェニル-9, 10-ビス((4-(2, 2-ジフェニルビニル)フェニル)アントラセン(化合物34)の合成

2-フェニル-9, 10-ビス(4-(2, 2-ジフェニルビニル)フェニル)-9, 10-ジヒドロキシ-9, 10-ジヒドロアントラセン(2.3g, 2.9mmol)を酢酸(20ml)に溶かし、よう化カリウム(1.4g, 8.4mmol, 3.6eq)、ホスフィン酸ナトリウム1水和物(0.4g, 3.8mmol)を加えて1時間攪拌した。反応混合物をろ別、水、メタノール、アセトンで洗浄して淡黄色固体(2.1g, 95%)を得た。

¹H-NMR(CDCl₃, TMS) δ 7.14(2H, s), 7.2-7.5(39H, m), 7.87(1H, s)

この固体(2.1g)を370 °C/10⁻⁶Torrで1時間昇華精製することにより淡黄色固体(0.9g)を得た。

FDMS, calcd for C₆₀H₄₂=762, found m/z=762(M⁺, 100)

λ_{max} 409, 388, 370nm(PhMe)

F_{max}, 453nm(PhMe, λ_{ex}=409nm)

E_g=2.85eV

I_p=5.70eV(14Y/eV, 100nW)

T_g=114°C

【0083】実施例1

25mm×75mm×1.1mm厚のITO透明電極付きガラス基板(ジオマティック社製)をイソプロピルアルコール中で超音波洗浄を5分間行なった後、UVオゾン洗浄を30分間行なった。洗浄後の透明電極ライン付きガラス基板を真空蒸着装置の基板ホルダーに装着し、まず透明電極ラインが形成されている側の面上に、前記透明電極を覆うようにして膜厚60nmのN,N'-ビス(N,N'-ジフェニル-4-アミノフェニル)-N,N-ジフェニル-4,4'-ジアミノ-1,1'-ビフェニル膜(以下、TPD232膜)を成膜した。このTPD232膜は、第一の正孔注入層(正孔輸送層)として機能する。次に、TPD232膜上に膜厚20nmの4,4'-ビス[N-(1-ナフチル)-N-フェニルアミノ]ビフェニル膜(以下、NPD膜)を成膜した。このNPD膜は第2の正孔注入層(正孔輸送層)として機能する。さらに、NPD膜上に膜厚40nmの化合物(15)を蒸着し成膜した。この膜は、発光層として機能する。この膜上に膜厚20nmのトリス(8-キノリノール)アルミニウム膜(以下、Alq膜)を成膜した。このAlq膜は、電子注入

層として機能する。この後Li (Li源: サエスゲッター社製)とAlqを二元蒸着させ、電子注入層(陰極)としてAlq: Li膜を形成した。このAlq: Li膜上に金属Alを蒸着させ金属陰極を形成し有機EL素子を作製した。この素子は直流電圧6Vで発光輝度80cd/m²、最大発光輝度23000cd/m²、発光効率2.0cd/Aの青色発光が得られた。さらに有機EL素子のスペクトルを図1に示す。同図に示すように、この有機EL素子はスペクトルがピーク波長450nm付近であり、色純度に優れていた。化合物(15)は、ガラス転移温度が118°Cと高耐熱であり、得られた有機EL素子を高温保存(85°C、500hr)したところ、性能に変化が無く、耐熱性に優れていることを確認した。また、初期発光輝度80cd/m²で、定電流駆動したところ半減寿命は13000時間と長寿命であった。

【0084】実施例2～13

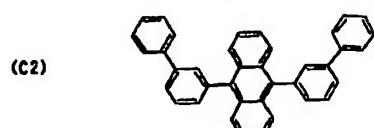
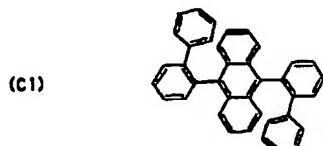
実施例1において、化合物(15)の代わりに表1に示す化合物を使用したことを除き同様にして、有機EL素子を作製し、直流電圧6Vで、発光輝度、発光効率を測定し、発光色を観察した。その結果を表1に示す。

【表1】

表 1

	化合物の種類	発光輝度(cd/m ²)	発光効率(cd/A)	発光色
実施例2	(1)	120	3.2	青
実施例3	(2)	113	2.7	青
実施例4	(3)	90	3.7	青
実施例5	(13)	130	2.2	青
実施例6	(14)	113	2.7	青
実施例7	(17)	90	4.2	青
実施例8	(20)	150	2.8	青
実施例9	(22)	180	4.7	青
実施例10	(25)	80	2.8	青
実施例11	(26)	75	2.6	青
実施例12	(33)	230	3.6	青
実施例13	(34)	280	4.3	青

【0085】実施例14～23及び比較例1～2
表2に示す本発明の新規化合物及び下記化合物(C1)
(比較例1)及び(C2)(比較例2)
【化25】



について、DSC測定によりガラス転移温度を測定した。その結果を表2に示す。

【0086】

【表2】

表 2

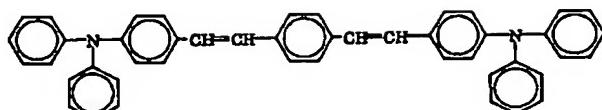
	化合物の種類	ガラス転移温度(℃)
実施例14	(14)	102
実施例15	(15)	118
実施例16	(17)	163
実施例17	(18)	106
実施例18	(22)	110
実施例19	(24)	113
実施例20	(25)	130
実施例21	(26)	135
実施例22	(33)	105
実施例23	(34)	110
比較例1	(C1)	75
比較例2	(C2)	97

表2に示したように、比較例の化合物が、ガラス転移温度100℃以下と耐熱性が低いのに対し、実施例14～23の化合物はガラス転移温度100℃以上の耐熱化合物であることが分かる。

【0087】実施例24

実施例1において、化合物(15)を蒸着する代わりに、化合物(15)及び下記化合物PAVB(蛍光性ドーパントとして機能する)

【化26】



を40:1の蒸着速度比で蒸着し製膜したこと以外は同様にして有機EL素子を作製し、直流電圧5.5Vで、発光輝度、発光効率及び最大発光輝度を測定し、発光色を観察した。その結果を表3に示す。

【0088】実施例25

実施例1において、化合物(15)を蒸着する代わりに、化合物(17)及び上記化合物PAVBを40:1の蒸着速度比で蒸着し製膜したこと以外は同様にして有機EL素子を作製し、直流電圧5.5Vで、発光輝度、発光効率及び最大発光輝度を測定し、発光色を観察し

た。その結果を表3に示す。

実施例26

実施例1において、化合物(15)を蒸着する代わりに、化合物(18)及び上記化合物PAVBを40:1の蒸着速度比で蒸着し製膜したこと以外は同様にして有機EL素子を作製し、直流電圧5.5Vで、発光輝度、発光効率及び最大発光輝度を測定し、発光色を観察した。その結果を表3に示す。

【0089】

【表3】

表 3

	化合物の種類	発光輝度(cd/m²)	発光効率(cd/A)	最大発光輝度(cd/m²)	発光色
実施例24	(15)とPAVB	222	7.14	85000	緑味青
実施例25	(17)とPAVB	135	7.58	75000	緑味青
実施例26	(18)とPAVB	145	9.67	95000	緑味青

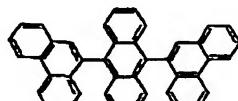
表3に示したように、本発明の新規化合物に蛍光性のドーパントを添加すると高効率になる。

【0090】比較例3

実施例1において、化合物(15)を蒸着する代わりに、下記化合物(C3)

【化27】

(C3)



を蒸着し製膜したこと以外は同様にして有機EL素子を作製した。この素子は直流電圧6.5Vで発光輝度92cd/m²、発光効率1.22cd/Aの青緑色発光であり、効率が低く実用的ではなかった。

【0091】実施例27

実施例1において、化合物(15)の代わりに化合物(25)を使用したことを除き同様にして、有機EL素子を作製した。この素子について、初期発光輝度500cd/m²で、定電流駆動したところ半減寿命は840時間(100cd/m²では、約6000時間に相当)と長寿命であり、高温保存(85°C、500時間)したところ性能に変化が無く、耐熱性に優れており、発光面の発光も均一で欠陥がなかった。また、この素子の発光効率は2.8cd/Aであり、色度座標(0.16, 0.08)と純度の高い青色発光が得られた。

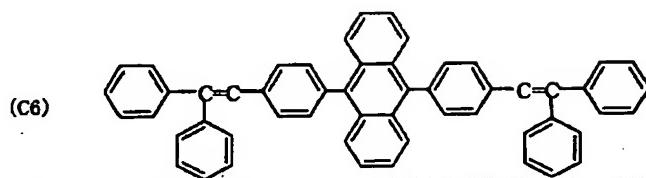
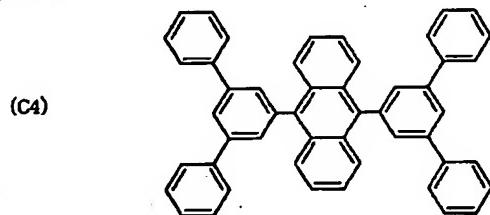
実施例28

実施例1において、化合物(15)の代わりに化合物(33)を使用したことを除き同様にして、有機EL素子を作製した。この素子について、初期発光輝度500cd/m²で、定電流駆動したところ半減寿命は1100時間(100cd/m²では、約8000時間に相当)と長寿命であり、高温保存(85°C、500時間)したところ性能に変化が無く、耐熱性に優れており、発光面の発光も均一で欠陥がなかった。また、この素子の発光効率は3.6cd/Aと優れていた。

【0092】比較例4

実施例1において、化合物(15)を蒸着する代わりに、下記化合物(C4)

【化28】



を蒸着し製膜したこと以外は同様にして有機EL素子を作製した。この素子について、初期発光輝度500cd/m²で、定電流駆動したところ半減寿命は1000時間と長いが、高温保存(85°C、500時間)したところ、発光面の一部に欠陥が発生し、欠陥部は、色が変化した。

【0095】以上のように、本発明の化合物を用いた素子は、発光効率が2cd/A以上の青色発光が実現可能で、比較例に対し優れている。さらに、長寿命、高耐熱であり、高温保存後も均一発光を維持できる。

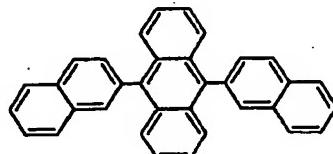
【0096】

を蒸着し製膜したこと以外は同様にして有機EL素子を作製した。この素子について、初期発光輝度500cd/m²で、定電流駆動したところ半減寿命は25時間と極めて短く、実用的ではなかった。また、発光効率は1.7cd/Aと低かった。

【0093】比較例5

実施例1において、化合物(15)を蒸着する代わりに、下記化合物(C5)

【化29】



を蒸着し製膜したこと以外は同様にして有機EL素子を作製した。この素子について、初期発光輝度500cd/m²で、定電流駆動したところ半減寿命は420時間と短く、実用的ではなかった。また、発光効率は2.1cd/Aと低かった。

【0094】比較例6

実施例1において、化合物(15)を蒸着する代わりに、下記化合物(C6)

【化30】

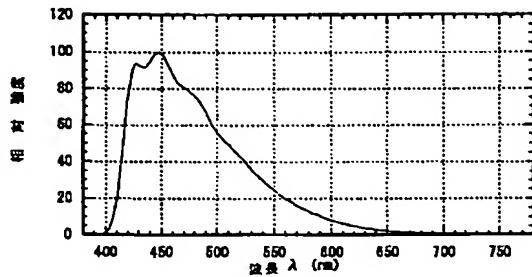
【発明の効果】以上、詳細に説明したように、上記[A]、[1]～[5]及び[3']で示されるいずれかの新規化合物を利用した本発明の有機エレクトロルミネッセンス素子は、発光効率が高く、耐熱性に優れ、寿命が長く、色純度が良い青色系に発光する。このため、本発明の有機エレクトロルミネッセンス素子は、壁掛テレビの平面発光体やディスプレイのバックライト等の光源として有用である。

【図面の簡単な説明】

【図1】 本発明の実施例1における有機エレクトロル

ミネッセンス素子のスペクトルを示す図である。

【図1】



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